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Bright electroluminescence in ambient conditions from WSe₂ p-n diodes using pulsed injection

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Transition metal dichalcogenide (TMDC) monolayers are promising for next-generation nanoscale light emitters due to their direct band gap, high optical efficiency, and ease of top-down fabrication. In this work, we study the time-resolved electroluminescence of monolayer WSe₂ lateral p-n junctions in ambient conditions, and identify the decay in current over time as the main issue preventing stable device operation. We show that pulsed voltage bias overcomes this issue and results in bright electroluminescence in ambient conditions. This is achieved in a simple back-gated transistor structure, without the use of dual gates, heterostructures, contact optimization, or chemical doping methods. Internal quantum efficiency (IQE) of electroluminescence reaches ~1%, close to the photoluminescence quantum efficiency, indicating efficient exciton formation with injected carriers. Emission intensity is stable over

27 **hours of device operation. Finally, our device exhibits ~15 ns rise**
28 **and fall times, to our knowledge the fastest direct modulation speed**
29 **reported for TMDC light-emitting diodes.**

30

31 I. INTRODUCTION

32 Monolayer transition metal dichalcogenides (TMDCs) are a class of single-
33 molecule-thick direct-bandgap semiconductors that show great potential for
34 next-generation electronics¹⁻⁴ as well as optoelectronic devices such as light-
35 emitting diodes (LEDs) and photodetectors⁵⁻¹². One long-recognized
36 challenge in TMDC electronics is the large hysteresis in TMDC field-effect
37 transistors (FETs). The cause of this hysteresis has been studied extensively,
38 with some results pointing to charge trapping caused by adsorption of water
39 and oxygen on the monolayer surface¹³⁻¹⁵, and others to intrinsic defects^{15,16}.
40 In electrically injected light-emitting devices, some form of electrostatic
41 gating is typically needed as well to form a P-N junction, due to the difficulty
42 of traditional doping techniques for monolayers, and so the same problem is
43 encountered. In FETs, hysteresis manifests as a shift in the I_d - V_g curve as
44 gate voltage is swept in opposite directions. At a constant gate bias, this is
45 seen as a decay in current over time^{14,15}. Although some past works on TMDC
46 electroluminescence have shown high EL quantum efficiencies $\sim 10^{-2}$, these
47 are reported only in high-vacuum conditions^{5,6,17,18}, where hysteresis is
48 partially mitigated.

49 A simple alternative approach is pulsed voltage bias, which has been shown
50 to yield hysteresis-free FET characteristics in 2D materials in ambient
51 conditions¹⁹⁻²¹. In addition, bright electroluminescence has been observed
52 from TMDC capacitors operated under fast pulsed bias¹². In such two-
53 terminal devices, electrons and holes are separately injected into the

54 semiconductor in alternating cycles (unipolar injection). In this work, we
55 show that pulsed injection is also effective for traditional bipolar injection
56 TMDC LEDs, yielding stable EL from monolayer WSe₂ devices in ambient
57 conditions. EL quantum efficiency is $\sim 10^{-2}$, close to that of
58 photoluminescence (PL). This implies that EL efficiency in our devices is
59 primarily limited not by charge carrier confinement but by intrinsic material
60 quality.

61 II. ELECTROLUMINESCENCE FROM WSe₂ P-N DIODES

62 Our device design (Fig. 1a) is a back-gated WSe₂ FET structure operated as a
63 p-n diode. The gate stack is either 50 nm SiO₂ on a p++ Si substrate, or 20
64 nm Al₂O₃ on a transparent ITO/glass substrate (see Methods). Devices on
65 both substrates showed bright light emission. WSe₂ was chosen due to its
66 ambipolar nature, required for bipolar carrier injection. In general, most
67 devices were ambipolar or slightly p-type in their I_d - V_g characteristics
68 (Supplementary Fig. S1, S7). Identical structures using monolayer MoS₂
69 showed only n-type I_d - V_g and no light emission (Supplementary Fig. S2). For
70 diode operation, one contact is designated as the P contact with alternating
71 voltage between V_p and V_g . The other (N) contact alternates between V_n and
72 V_g , with $V_n < 0$ (Fig. 1b). Light is emitted only during the on period t_{on} . Fig. 1c
73 shows the EL spectrum of the device overlaid with the photoluminescence
74 (PL) spectrum of the same device. The clear peak at ~ 1.65 eV shows that EL
75 and PL are both due to the usual recombination of A excitons, and confirms
76 the monolayer nature of the flakes. PL quantum efficiency versus pump

77 intensity shows a drop at high pump intensity due to exciton-exciton
78 recombination, similar to past work²² (Supplementary Fig. S3). Fig. 1d shows
79 the light emission overlaid on an image of the device, confirming emission
80 comes from the channel region.

81 To illustrate the need for pulsed bias, we measure EL versus time and
82 current versus time for both DC (step) voltage and pulsed voltage at 5 kHz.
83 Bright emission is observed under both DC and pulsed bias. Note that the
84 emission mechanism in our devices is clearly distinct from the pulsed light-
85 emitting capacitor reported previously¹², since continuous light emission is
86 seen on a scale of ~seconds, as opposed to the ~10 ns pulses that only
87 occur during voltage transitions. However, under DC bias, both emission and
88 current rapidly decay by orders of magnitude within a few seconds (Fig.
89 2a,b). Past reports on current decay in MoS₂ FETs show roughly comparable
90 time constants ~10 s^{14,15}. The ratio of light emission to current, which is
91 proportional to the efficiency, remains roughly constant over time, showing
92 that the decay in current is responsible for the decay in light emission. In
93 contrast, pulsed bias yields extremely stable light emission and current over
94 >1000s (Fig. 2c,d). Most devices under pulsed bias showed no decrease or
95 small decrease (<2x) in light emission in the first few minutes, then stayed
96 stable for the remaining duration of the applied bias, with the longest test
97 performed being >3 hours (Supplementary Fig. 4). The frequency response is
98 shown in Fig. 2e. Here, emission intensity is defined as the average intensity
99 over 10s, starting 5s after the pulsed bias is applied. Above ~1 kHz, emission

100 intensity is relatively stable with frequency, while below ~ 1 kHz, current
101 decay causes EL to drop very quickly. However, note that the stability also
102 depends on duty cycle, which is fixed at 50% here. For very low duty cycle
103 (long t_{off}), emission intensity can still be stable at low frequencies ~ 1 Hz
104 (Supplementary Info).

105 Next, we study the emission characteristics under varying bias to extract the
106 optimal bias condition. The relative injection level of electrons and holes, and
107 thus the exciton formation efficiency, depends on the voltages ($V_p - V_g$) and
108 ($V_n - V_g$) applied to the contacts. If $|V_p - V_g| \gg |V_n - V_g|$, holes will be
109 predominantly injected and will simply diffuse across the channel to the
110 opposite contact without forming excitons, and vice versa for
111 $|V_p - V_g| \ll |V_n - V_g|$ (Fig. 3a). We determine the optimal bias condition by
112 keeping ($V_p - V_n$) constant and sweeping V_g from V_n to V_p while tracking the
113 EL and current. Fig. 3b shows the EL intensity versus gate voltage, where V_p
114 and V_n are fixed at $V_p = 5V$ and $V_n = -5V$, along with the current and EL
115 efficiency. Each point corresponds to one on period $t_{on} = 0.5s$, and current and
116 emission intensity are defined as the time-average during the on period. An
117 off time of $t_{off} = 10s$ is used to recover the device between pulses
118 (Supplementary Info). The brightest emission occurs near $V_g = 0$, where both
119 holes and electrons are injected. The minimum current point at $V_g \approx 1.8V$
120 corresponds to the most bipolar injection, and coincides with the highest

121 internal quantum efficiency (IQE) $\sim 1\%$. $|V_p - V_g| < |V_n - V_g|$ at this point,
122 indicating slightly p-type characteristics typical of monolayer WSe₂. We
123 obtain an L-I curve at optimal bias conditions by first setting $V_g = 1.25$ V,
124 approximately the point of maximum efficiency above, where current is also
125 not too low. Then we increase the source-drain bias while keeping
126 $|V_p - V_g| < |V_n - V_g|$ constant at ~ 0.6 . For comparison, we also take an L-I
127 curve with $V_g = 0$ (i.e. equal $|V_p - V_g|$ and $|V_n - V_g|$). L-I and IQE are
128 plotted in Fig. 3c, along with the corresponding PL L-L curves. PL shows a
129 roughly constant IQE of $\sim 1.6\%$, while EL at optimal bias shows a peak of
130 $\sim 1.3\%$ at low current and decreases slightly at higher currents, possibly due
131 to variation in optimal bias condition with voltage. In contrast, EL at equal P
132 and N bias is low ($\sim 0.3\%$) at low current and stays below $\sim 0.8\%$ throughout.

133 Next, we study the modulation speed of the device by performing time-
134 resolved measurements using time-correlated single-photon counting
135 (TCSPC). Supplementary Fig. S5 shows the measurement setup, and Fig. 4
136 shows time-resolved electroluminescence at 1 MHz, together with the P and
137 N voltage pulses measured on an oscilloscope. Emission only occurs when
138 both P and N voltage are applied, confirming bipolar carrier injection. The
139 emission intensity is nearly constant during the entire P/N voltage overlap
140 period. The rise and fall times shown in the inset are ~ 12 ns and ~ 18 ns
141 respectively. We note that this rise/fall time is $\sim 20\times$ faster than monolayer
142 LEDs using vertical tunnel injection heterostructures²⁴, which is likely due to

143 the lower capacitance of the lateral injection scheme used here. To our
144 knowledge, this is the fastest direct modulation speed reported for TMDC
145 LEDs.

146 I. CONCLUSION

147 In this work, we have shown that the hysteresis and current decay commonly
148 seen in TMDC transistors also play an important role in light-emitting diodes.
149 Pulsed injection is an effective way to circumvent this issue, yielding bright
150 and stable EL using a simple back-gated FET structure, with efficiency near
151 that of PL. We show how to extract the optimal bias condition for efficient
152 bipolar injection, and study the high frequency behavior of light emission. A
153 fast ~ 15 ns rise/fall time is observed, indicating strong potential for high
154 speed light modulation. Pulsed emission is stable over hours of operation.
155 Further improvements in efficiency will come from contact optimization to
156 enable lower voltage operation, as well as advances in CVD growth to
157 improve intrinsic material quality. Higher speed can be obtained by coupling
158 to an optical cavity to enhance spontaneous emission rate²⁴⁻²⁷.

159 II. METHODS

160 Device fabrication for SiO₂/Si substrates begins with a p++ Si substrate with
161 50 nm thermal oxide (Silicon Valley Microelectronics), with alignment marks
162 (5/60 nm Ti/Au) deposited by electron beam evaporation. Monolayer WSe₂
163 flakes are grown on quartz using chemical vapor deposition (CVD) using
164 similar conditions as in Ahn *et al*²⁸. Flakes ~ 100 μm in size are transferred

165 onto the gate oxide using a pick-and-place method, and source/drain
166 contacts are patterned using e-beam lithography (EBL) followed by thermal
167 evaporation (15 nm Ni) and liftoff in acetone for 30 min. Devices on
168 transparent substrate use ITO (280 nm)/glass (Thin Film Devices) with 20 nm
169 Al_2O_3 deposited by atomic layer deposition. Before contact deposition, flakes
170 are patterned with EBL and etched in XeF_2 (3T XeF_2 , 4T N_2 , 30 s/cycle, 3
171 cycles).

172 Electroluminescence measurements are carried out with a low-frequency
173 probe setup using two Keithley 2401 SourceMeters for quasi-DC
174 measurements, or two synchronized HP33120A function generators for
175 TCSPC and frequency response. For quasi-DC and frequency response
176 measurements, light is collected from the top through a 20x objective and
177 focused onto either a grating for spectral measurement, or mirror for spatial
178 measurement, then onto a Si CCD. For TCSPC measurements, light is
179 collected through the backside with a 20x objective and focused onto a Si
180 avalanche photodiode (Micro Photon Devices) synchronized with a trigger
181 pulse from a DG535 pulse generator (Stanford Research Systems).
182 Photoluminescence measurements are taken using the same microscope
183 setup as the quasi-DC tests, with a 532 nm pump laser focused onto the
184 flake using a 50x objective.

185 Electrical measurements are done using an Agilent B1500 parameter
186 analyzer or a HP4145b parameter analyzer. Vacuum measurements are done

187 using a Lakeshore TTPX probe station. Gas response measurements are done
188 using a home built gas sensing chamber.

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199 IV. AUTHOR CONTRIBUTIONS

200 K.H. and M.C.W. designed the study. K.H. performed the measurements and
201 analysis, and wrote the manuscript. G.H.A., J.C., and G.Z. performed the CVD
202 growth. M.A. helped with electrical measurements. D.-H.L. and S.B.D.
203 provided helpful discussion. H.K. helped with MoS₂ exfoliation. N.G. helped
204 with gas sensing measurements. A.J. advised on the experiments and
205 manuscript.

206

207 V. REFERENCES

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280

281

Figure Captions

Figure 1. Light emission from pulsed WSe₂ p-n diodes. **(a)** Device schematic. Contact/gate oxide/gate stack is either Ni (15 nm)/SiO₂ (50 nm)/Si or Ni (15 nm)/Al₂O₃ (20 nm)/ITO. **(b)** Schematic of voltage pulsing. Light is emitted when both $V_p > V_g$ and $V_n < V_g$, while during the off state $V_p = V_n = V_g$. **(c)** EL and PL spectra taken on same device. **(d)** Spatial map of emission from an etched device. Left: optical micrograph. Scale bar is 5 μ m. Middle: emission intensity during pulsed injection, overlaid on CCD image. Emission occurs in the channel region between the two contacts. Right: device in off state with no bias applied.

Figure 2. Light emission and current for DC and pulsed bias. **(a)** Schematic of DC voltage bias. **(b)** Light emission and current over time for DC voltage of $V_p = -V_n = 4V$, with $V_g = 0V$. **(c)** Schematic of pulsed voltage bias, using a square wave with 50% duty cycle. **(d)** Light emission and current over time for pulsed voltage with $V_p = -V_n = 4V$, with $V_g = 0V$. Note the different time scale for (b) and (d). **(e)** Frequency response with $V_p = -V_n = 4.5 V$, taken with a different device from (b, d).

Figure 3. L-I-V characteristics and optimal bias condition. **(a)** Schematic of carrier injection regimes with varying gate voltage and fixed V_p and V_n . **(b)** EL

304 intensity, current, and EL IQE versus gate voltage V_g , for constant $V_p = -V_n =$
305 5 V. (c) Output power and IQE vs carrier injection rate for PL and EL. EL is
306 taken at near-optimal bias condition extracted from V_g sweep ($V_g = 1.25$ V),
307 as well as $V_g = 0$ V for comparison.

308

309 Figure 4. Time-resolved light emission. Top panel: Voltage pulses measured
310 on an oscilloscope. Bottom panel: Time-resolved EL. Left inset: close-up of EL
311 rise time. Right inset: close-up of EL fall time.

312 **Supplementary Info**

313 *Causes of current decay in ambient conditions*

314 To investigate the cause of the current decay, we studied the device in
315 vacuum (3×10^{-5} Torr) and after annealing at 140 C for 2 hours to remove
316 adsorbates, followed by cooling back to room temperature. The current
317 under electron injection ($V_{ds} = 1V$, $V_g = 8V$) becomes highly stable while the
318 hole current ($V_{ds} = 1V$, $V_g = -10V$) still decays (Supplementary Fig. 6a). In
319 ambient conditions for the same device, both electron and hole currents
320 decay similarly (Supplementary Fig. 6b). In addition, the I_d/V_g curve shows
321 decreased hysteresis in vacuum, and a shift in threshold voltage for
322 electrons but not for holes (Supplementary Fig. 7). Therefore, we attribute
323 the current decay in ambient to both adsorbed molecules on the surface of
324 the monolayer, as well as additional hole traps that remain after removing
325 adsorbates, possibly due to interface traps or intrinsic defects^{16,23}. This shows
326 that pulsing is still necessary in vacuum to obtain stable bipolar current
327 injection. In air, H_2O and O_2 are the primary constituents possibly responsible
328 for current decay¹³. To test the relative contributions of H_2O and O_2 on the
329 device, we measured hysteresis in ambient air, dry air (21% O_2 , 79% N_2), and
330 pure N_2 (Supplementary Fig. 8). The hysteresis for both electrons and holes
331 is higher in ambient than the other cases, while almost identical for dry air
332 and pure N_2 . Therefore, adsorbed water is likely the primary factor affecting
333 the device in ambient conditions, similar to MoS_2 FETs^{13,14}.

335 Calculation of internal quantum efficiency (IQE)

336 For electroluminescence, IQE is determined by the formula $\eta_{EL} = \frac{P}{\eta_{setup}\eta_{extr}I}$,

337 where P is the output photons/s collected, I is the input carriers/sec, η_{setup} is

338 the efficiency of the optical setup, including objective, focusing optics,

339 spectrometer, and CCD, and η_{extr} is the extraction efficiency from the

340 substrate. η_{setup} is measured using a 532 nm laser incident on a spectralon

341 sample to simulate an ideal Lambertian emitter, which then goes through the

342 same optics as the EL measurement. This gives the absolute setup efficiency

343 at 532 nm. Then, a blackbody source with known spectrum is shone on the

344 sample and used to find the relative spectral efficiency, in particular the

345 efficiency at 750 nm, the wavelength of EL. We obtain $\eta_{setup} = 4.3 \times 10^{-3}$. η_{extr}

346 is estimated from FDTD simulation of monolayer WSe₂ on SiO₂/Si substrate to

347 be ~ 0.1 . For photoluminescence, the efficiency is also divided by the

348 absorbance A of WSe₂ at 532 nm. This is estimated from FDTD simulation to

349 be 13.7% on SiO₂/Si.

350

351 *Emission decay for varying off periods*

352 For quasi-DC measurements with on period of 0.5s, the off period should be
353 sufficiently long to avoid current decay between pulses. Off periods between
354 0.2s and 10s were tested with a sequence of 10 pulses for each off period.
355 The device was rested for 30s after each 10 pulse sequence. Emission
356 intensity is plotted in Supplementary Fig. S9. For pulse periods below ~ 0.5 s,
357 a clear decay is seen, while with the 10s rest period used for the V_g sweep
358 measurements, the intensity fluctuates but there is no decay trend.