Supplementary Materials for

Injection-free multiwavelength electroluminescence devices based on monolayer semiconductors driven by an alternating field

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Section S1. Device Fabrication

The fabrication process of our devices mainly includes three steps. Firstly, we prepare the interdigital electrodes (IDEs) on \(\text{SiO}_2/\text{Si}\) substrates through electron beam (EB) lithography, electrodes deposition (30 nm/50 nm, Cr/Au), and lift-off (Fig. S1a). Secondly, the monolayer transition metal dichalcogenides (TMDCs) and hexagonal boron nitride (h-BN) flakes are mechanically exfoliated from bulk materials (supplier: HQ Graphene and 2D Semiconductors) and covered onto polydimethylsiloxane (PDMS)/glass-slide substrates, respectively. The transparent PDMS/glass-slide substrates are selected for alignment during the following transfer process. Finally, the h-BN flakes and monolayer TMDCs are transferred onto the IDEs (Fig. S1b, c) by the dry transfer technique. An h-BN flake as an insulating and dielectric layer is firstly transferred onto the IDEs, with the thickness in the range of 20-50 nm. Then, the monolayer TMDCs and a cap h-BN flake are vertically stacked by repeating the transfer process. The dry transfer process is carried out on a homemade micromanipulator with a microscope.

Figure S1. Optical images of a device on each step of fabrication. a, IDEs/\(\text{SiO}_2\) substrate. b, Transferring h-BN layer formed h-BN/IDEs/\(\text{SiO}_2\) structure. c, Transferring TMDC materials formed TMDC/h-BN/IDEs/\(\text{SiO}_2\) structure. d, The dark field image of our device.
Section S2. Optical and Electrical Measurement

The measurement setup for the micro-photoluminescence (PL) and electroluminescence (EL) includes a homemade microscope and a spectrometer equipped with a liquid nitrogen-cooled Si 2D detector and a thermoelectric cooled InGaAs 2D detector. The PL and EL spectra and EL imaging of the devices based on monolayer WSe$_2$, WS$_2$, MoSe$_2$, and MoS$_2$ are detected by Si detector, while those of the devices based on monolayer and bilayer MoTe$_2$ are detected by InGaAs detector. The PL and EL spectra and EL imaging of the devices based on monolayer WSe$_2$, WS$_2$, MoSe$_2$, and MoS$_2$ are pumped by a continuous-wave (CW) semiconductor laser at 532 nm. The PL of monolayer MoTe$_2$ is pumped by a CW He-Ne laser at 633 nm. The pumped light is vertically incident onto the sample plane by a ×100 (or ×20) objective with numerical aperture NA=0.7 (or 0.4), which also collects the emission light. In EL measurements, the devices are driven by alternating current (AC) voltages generated from a semiconductor parameter analyzer (B1500A, Keysight), and the emitting light is collected using the measurement setup mentioned above. EL measurements at low temperatures as shown in Fig. S7 are operated in a vacuum in a cryostat, while the EL measurements at room temperature are operated in ambient conditions.

The time-resolved light pulses are detected by a time-correlated single-photon-counter (TCSPC) integrated into the micro-PL setup. The trigger for TCSPC is also generated from B1500A, possessing the same frequency and duty cycle (50%) with AC trigger voltage for device operation. The result (Fig. S2a) shows that the driving and trigger voltages have the same phase, so the 200 ns delay of light pulses after driving voltage switching (Fig. 1e) is mainly caused by the resistance-capacitance effect of the device. The capacitance of the device is about 1 nF below 0.5 MHz (Fig. S2b), measured with Keysight B1500A. The AC resistance of the device is estimated at about 185 Ω, using the formula: $U_d/U_s = 1/(1+j\omega RC)$, where $U_d$ is the effective voltage on the device and $U_s$ is source voltage, $C$ is capacitance and $R$ is AC resistance, $\omega = 2\pi f$, $f = 0.25$ MHz here.

In order to investigate the possibility of dielectric breakdown of the h-BN layer in our device or carrier tunneling within the voltage range applied in the experiment, we test the AC current of the device under an alternating square wave voltage. The measurement circuit is shown in Fig. S2c. The device is connected in series with an alternating voltage source and a resistance ($R$) of 10 Ohms. $V_1$ and $V_2$ show the voltages applied on the IDEs and the resistance ($R$), measured by the oscilloscope. Figure S2d shows the time-synchronized alternating voltage and current across the device. The peak voltage between IDEs is 42 V and the period is 4 µs. It can be seen from Fig. S2d that the peak value of the alternating current appears at the edge of the voltage change. When the voltage is stable, the current drops to zero. This is the voltage-current characteristic of the typical capacitor charging and discharging process. If there is breakdown or tunneling of h-BN, the leakage current will be very large in the whole period at high voltages. Therefore, the voltage-current characteristic of Fig. S2d shows that there is no breakdown or tunneling of h-BN, and there is no carrier exchange between IDEs and TMDCs.
Figure S2. The electrical characteristics of the IDEs based device. **a**, The synchronous of the AC voltage for device driving (black line) and TCSPC trigger (red line). **b**, Device capacitances under different frequencies. **c**, The circuit of alternating current measurement of the device. **d**, Time-synchronized voltage and alternating current across the device.

**Section S3. EL Spectrum Fitting**

An EL spectrum of a monolayer WSe₂ based device is fitted with Voigt lineshape. The red line with a peak at 759 nm represents the emission from excitons. The blue line with peak at 783 nm represents the emission from trions.

Figure S3. Fitting curves of an EL spectrum of a monolayer WSe₂ based device.
### Section S4. System Collection Efficiency Calibration

We calibrate the overall measurement efficiency, defined as $\eta = \eta_{\text{sys}}\eta_{\text{lens}}$. $\eta_{\text{sys}}$ is transmission efficiency of the measurement setup, including the efficiency of all optical components, slit, grating, and Si CCD. We directly measure $\eta_{\text{sys}}$ by taking spectra of the 780 nm laser that reflected from a silver mirror. We assume that the value of $\eta_{\text{sys}}$ at 780 nm is approximately the same as those from 600 nm to 800 nm, which covers the emission wavelengths of monolayer WS$_2$, MoS$_2$, WSe$_2$, and MoSe$_2$. The power of the 780 nm laser incident to the silver mirror is 0.5 nW ($1.96\times10^9$ cts/sec), while the reflected light into CCD is $1.51\times10^8$ cts/sec. Therefore, we obtain an estimate of $\eta_{\text{sys}}$ for the measurement setup used to measure devices 1#-4# to be $\eta_{\text{sys}} = 1.51\times10^8/1.96\times10^9 = 0.077$, while for device 5# and 6# is $\eta_{\text{sys}} = 0.047$ (Table S2).

The lens collection efficiency, $\eta_{\text{lens}}$, depends on the NA of the objective and the radiation field distribution. For isotropic emission, $\eta_{\text{lens}}$ is usually defined as a ratio of the integral over the maximum collection angle ($\theta_M$) over the whole solid angle of $4\pi$, namely

$$
\eta_{\text{lens}} = \frac{\int_0^{2\theta_M} \int_0^{\alpha} \sin \theta d\theta d\phi}{4\pi} = \frac{2\pi (1-\cos \theta_M)}{4\pi} = \frac{1}{2} \frac{1}{2} \cos \theta_M
$$

(S1)

where, $\theta_M$ is given by NA and the refractive index of the material ($n_r$) covered on top, given as $\theta_M = \arcsin(\text{NA}/n_r)$.

For the Lambertian sources, the expression of $\eta_{\text{lens}}$ is

$$
\eta_{\text{lens}} = \frac{\int_0^{2\theta_M} \int_0^{\pi/2} \cos \theta \sin \theta d\theta d\phi}{2\pi \int_0^{\pi/2} \cos \theta \sin \theta d\theta d\phi} = \frac{1}{2} \frac{1}{2} \cos^2 \theta_M
$$

(S2)

Different from the isotropic distribution of dipoles in the conventional bulk cubic materials, emission of 2D monolayer materials originates mostly from dipoles oriented in the plane of 2D layers, at least in the range of emission energy near bright excitons. In addition, the entire sample structures of 2D-material-based devices are much thinner than emission wavelengths, multiple reflections and total internal reflection can be ignored. Therefore, the emission of 2D materials can be considered as in-plane dipoles emitting directly to $4\pi$ solid angle. In our device, the emission pattern is treated as symmetric with respect to Y to -Y inversion. If we assume Z-X is the plane of 2D monolayer and dipoles are along the Z-axis as shown in Fig. S4, the value of $\eta_{\text{lens}}$ could be estimated by integrating the dipole emission profile over $\theta_M$. 

Figure S4. Schematic diagram of the dipole emission and objective collection, where the thick red bar indicates the dipole.

The time-averaged power radiated per unit solid angle is given by (45)

\[
\frac{dP}{d\Omega} = \frac{c^2 Z_0}{32\pi^2 k^4} |\mathbf{p}|^2 \sin^2 \theta = P_0 \sin^2 \theta,
\]

where \(\theta\) is the angle of measurement direction with respect to the orientation of dipole (Z-axis). For the convenience of calculating the solid angle around the objective direction (Y), we use transformation between \((\theta, \phi)\) and \((\theta', \phi')\):

\[
\begin{align*}
x &= r \sin \theta \cos \phi = r \sin \theta' \sin \phi' \\
y &= r \sin \theta \sin \phi = r \cos \theta' \\
z &= r \cos \theta = r \sin \theta' \cos \phi'
\end{align*}
\]

Therefore, \(\sin^2 \theta = 1 - \sin^2 \theta' \cos^2 \phi'\).

The collected power by an objective along Y-axis can be expressed as

\[
P_c = P_0 \int_0^{2\pi} \int_0^{\theta_M} \sin^2 \theta \sin \theta' \, d\theta' \, d\phi' = P_0 \int_0^{2\pi} \int_0^{\theta_M} \left(1 - \sin^2 \theta' \cos^2 \phi'\right) \sin \theta' \, d\theta' \, d\phi'
\]

\[
= P_0 \left[ \int_0^{2\pi} \int_0^{\theta_M} \sin \theta' \, d\theta' \, d\phi' - \int_0^{2\pi} \int_0^{\theta_M} \sin^3 \theta' \cos^2 \phi' \, d\theta' \, d\phi' \right]
\]

\[
= P_0 \left[ 2\pi - 2\pi \left( \frac{1}{3} - \frac{1}{2} \cos \theta_M + \frac{1}{6} \cos^3 \theta_M \right) \right]
\]

\[
= 2\pi P_0 \left[ \frac{2}{3} - \frac{1}{2} \cos \theta_M - \frac{1}{6} \cos^3 \theta_M \right]
\]
The total dipole emission power is \( P_r = P_0 \int_0^{2\pi} \int_0^\theta \sin^2 \theta \sin \theta' \sin \theta' d\theta' d\varphi' = 2\pi P_0 \left( \frac{4}{3} \right) \). The lens collection efficiency for the in-plane dipoles is

\[
\eta_{\text{lens}} = \frac{P_c}{P_r} = \frac{1}{2} \left( 1 - \frac{3}{8} \cos \theta_M - \frac{1}{8} \cos^3 \theta_M \right). \tag{S3}
\]

The values of \( \eta_{\text{sys}}, \eta_{\text{lens}} \) (adopting Eq. S3), and overall measurement efficiency (\( \eta \)) of the setup used for devices 1#-6# are listed in Table S2.

**Table S1. Comparisons of \( \eta_{\text{lens}} \) and conversion efficiency (CE) between two light emission cases**

| Maximum collection angle (\( \theta_M \)) | Isotropic emission (Eq. S1) | In-plane dipoles in 2D (Eq. S3) |
|------------------------------------------|-------------------------------|---------------------------------|
| \( \theta_M = 0.324, \text{RT} \)       | \( \eta_{\text{lens}} \)     | \( \eta_{\text{lens}} \)       |
|                                          | 0.026                         | 0.038                           |
|                                          | 23.3\%                        | 16\%                            |

Note: RT = room temperature.

**Table S2. \( \eta_{\text{sys}}, \eta_{\text{lens}}, \) and \( \eta \) of the measurement setup for devices 1#-6#**

|                                 | Device 1#-4# | Device 5#, 6# |
|--------------------------------|--------------|---------------|
| \( \eta_{\text{sys}} \)       | 0.077        | 0.047         |
| \( \eta_{\text{lens}} \) (for in-plane dipoles) | 0.038 | 0.012 |
| \( \eta = \eta_{\text{sys}} \eta_{\text{lens}} \) | \( 2.9 \times 10^{-3} \) | \( 5.6 \times 10^{-4} \) |

Section S5. Simulation of the Electric Field

The electric field distribution and intensity are calculated by the finite element method (FEM) in steady state, neglecting the screen effect causing by carriers considering the low carrier density. In our devices, monolayer TMDCs are usually placed on the IDEs with a distance in the range of 20-50 nm. As shown in Fig. S5a, the red arrows represent the electric field lines, and the color bar is in the log scale. Figure S5b shows the electric field (\( F \)) intensity along the X direction at the position of 20 nm above the surface of IDEs (Y=70nm) with different bias voltages (\( V \)), which indicate the electric intensity in the monolayer TMDCs of our devices.
Figure S5. Simulated electric field intensity between IDEs. a, The simulation of electric field intensity distribution and electric field line between two gates. b, In-plane electrical field intensities with different bias voltages ($V$) on the position of 20 nm above the IDEs. The electric field intensities in the center of the gap were chosen as the average values in the active material.

Section S6. Modeling of Impact Excitation, Light Emission Processes, and Conversion Efficiency

The entire physical processes in the impact region are depicted in Fig. 2a. The excitons are generated by impact excitation, and then radiatively or non-radiatively recombined or dissociated in the strong electric field. The electrons and holes from dissociated excitons can participate in the secondary impact excitation to create new excitons or drift outside of the acceleration region. We describe the physical processes mentioned above with rate equations, as follows:

\[
\frac{dN}{dt} = \Gamma_e n - \Gamma_d N - (\gamma_r + \gamma_{nr}) N
\]

(S4)

\[
\frac{dn}{dt} = J + \Gamma_d N - \gamma_d n
\]

(S5)

where $N$ is exciton areal density, $n$ is free carrier areal density, $\Gamma_e$ is impact excitation rate, $\Gamma_d$ is dissociation rate of an exciton, $\gamma_r = 1/\tau_r$ is exciton radiative recombination rate, $\gamma_{nr} = 1/\tau_{nr}$ is the non-radiative recombination rate, $\gamma_d$ is the rate for carriers to drift out of impact region, and $J$ is carrier flux density that injected into impact region.

In steady-state, the exciton density $N$ and carrier density $n$ satisfy:

\[
\frac{dN}{dt} = \frac{dn}{dt} = 0
\]

Solving the equations yields the steady-state solution:

\[
N = \frac{J\Gamma_e}{\Gamma_d (\gamma_d - \Gamma_e) + (\gamma_r + \gamma_{nr}) \gamma_d}
\]

(S6)
\[ n = \frac{J (\Gamma_e + \gamma_e + \gamma_m)}{\Gamma_d (\gamma_d - \Gamma_e) + (\gamma_e + \gamma_m) \gamma_d} \quad (S7) \]

The impact excitation rate \((\Gamma_e)\) means exciton generation by impact excitation per second, given by (34):

\[ \Gamma_e = A \exp\left(-F_e / F\right) \quad (S8) \]

where \(A\) is a constant coefficient, which represents the saturated rate of \(\Gamma_e\). \(F_e\) is the critical electric field intensity. \(F_e\) roughly obeys the relationship (34) \(F_e = 1.5E_x/(e\lambda_m)\), where \(E_x\) is exciton energy, \(e\) is the elementary charge, and \(\lambda_m\) is mean free path (MFP) of carriers. The exciton dissociation rate \((\Gamma_d)\) is given by (46):

\[ \Gamma_d = B \exp\left(-F_d / F\right) \quad (S9) \]

where \(B\) is a constant coefficient, \(F_d\) is the critical electric field intensity. \(F_d\) is associated with binding energy \((E_b)\) of an exciton, given by (46) \(F_d = E_b/(ed)\), where \(d\) is exciton Bohr radius, \(e\) is the elementary charge. Carriers drift-out rate \(\gamma_d = 2\mu F/W\), where \(W\) is the width of impact region, and \(\mu\) is carrier mobility.

The integrated emitted photon number in a single pulse duration \((T_{\text{pulse}})\) can be expressed as:

\[ N_{ph0} = \int_0^{T_{\text{pulse}}} S N \gamma_e dt = \int_0^{T_{\text{pulse}}} S \gamma_e \frac{J \Gamma_e}{\Gamma_d (\gamma_d - \Gamma_e) + (\gamma_e + \gamma_m) \gamma_d} dt \]

where \(S\) is the area of the active region. Then we obtain:

\[ N_{ph0} = \frac{\gamma_e \Gamma_e}{\Gamma_d (\gamma_d - \Gamma_e) + (\gamma_e + \gamma_m) \gamma_d} \int_0^{T_{\text{pulse}}} S J dt = N_c \frac{\gamma_e \Gamma_e}{\Gamma_d (\gamma_d - \Gamma_e) + (\gamma_e + \gamma_m) \gamma_d} \]

where \(N_c = \int_0^{T_{\text{pulse}}} S J dt\) is the number of overall pre-existing free carriers in the active material. In the total integration time \((T)\) of the detector, the pulse number is \(n_{\text{pulse}} = 2T/T_e\), where \(T_e\) is the period time of square wave driving voltage and factor 2 reflects the fact that there are two light pulses generated in a cycle of square wave voltage. Taking into account the overall measurement efficiency, \(\eta\), as defined in Section S4, we obtain the total number of measured photons within the integration time of \(T\), \(N_{ph}\), as given by

\[ N_{ph} = \eta n_{\text{pulse}} N_{ph0} = \eta n_{\text{pulse}} N_c \frac{\gamma_e \Gamma_e}{\Gamma_d (\gamma_d - \Gamma_e) + (\gamma_e + \gamma_m) \gamma_d} \quad (S10) \]

Although our devices operate in alternating mode and light emission is pulsed, considering the width of a single light pulse is about several tens of nanoseconds, much longer than impact excitation, dissociation and recombination processes (~ps), the steady-state solution is reasonable for physical model description and data fitting as plotted in Fig. 2c in the main text. The parameters utilized for fitting are listed in Table S3, and the fitting
results are listed in Table S4.

Table S3. Parameters utilized for fitting

| Parameters               | 10 (WS₂) | 2# (MoSe₂) | 3# (MoSe₂) | 4# (WS₂) | 5# (WS₂) | 6# (WS₂) |
|--------------------------|----------|------------|------------|----------|----------|----------|
| \( E_e \) (eV)           | 2        | 1.57       | 1.57       | 1.65     | 1.65     | 1.65     |
| \( d \) (nm)             | 1        | 1          | 1          | 1        | 1        | 1        |
| \( \eta \)               | 2.9×10³  | 2.9×10³    | 2.9×10³    | 2.9×10³  | 5.6×10⁴  | 5.6×10⁴  |
| \( S \) (µm²)            | 50       | 175        | 500        | 510      | 150      | 200      |
| \( n_0 \) (cm⁻³)         | 1×10¹⁰   | 1×10¹²     | 1×10¹²     | 1×10¹⁰   | 1×10¹⁰   | 1×10¹⁰   |
| \( W \) (µm)             | 0.4      | 0.4        | 0.4        | 1        | 0.4      | 0.4      |
| \( T \) (s)              | 5        | 3          | 5          | 10       | 5        | 5        |
| \( T_r \) (µs)           | 40       | 10         | 1          | 100      | 4        | 4        |

Note: \( n_0 \) is measured by the four-probe method in FET devices as described in Section S8.

Table S4. Fitting results

| Parameters               | 10 (WS₂) | 2# (MoSe₂) | 3# (MoSe₂) | 4# (WS₂) | 5# (WS₂) | 6# (WS₂) |
|--------------------------|----------|------------|------------|----------|----------|----------|
| \( A \) (s⁻¹)            | 6.5×10¹¹ | 3×10¹⁰    | 9×10¹⁰    | 4×10⁹   | 4×10¹⁷   | 2×10¹⁰   |
| \( \lambda_m \) (nm)     | 7.3      | 6.5        | 5.2        | 11       | 4.1      | 14       |
| \( \mu \) (cm²V⁻¹s⁻¹)    | 1×10⁵⁷   | 3×10¹¹     | 3×10¹¹     | 3×10¹¹   | 1×10¹⁰   | 8×10¹⁰   |
| \( E_b \) (meV)          | 220      | 260        | 300        | 400      | 380      | 400      |
| \( \tau_{nr} \) (ps)     | 0.1      | 0.4        | 0.6        | 0.08     | 0.03     | 0.09     |
| \( \tau_r \) (ms)        | 30       | 90         | 18         | 80       | 32       | 18       |
| \( \eta \)               | 1        | 3          | 0.6        | 2        | 0.8      | 0.3      |
| PLQY                     | 3%       | 3%         | 3%         | 4%       | 4%       | 6%       |

The fitting results of \( E_b \) are in reasonable range, \( \lambda_m \) are consistent with the values in reference(47), \( \mu \) is consistent with the values from our own FETs measurement as plotted in Fig. S6d. \( \tau_{nr} \), \( \tau_r \) and derived PLQY (= \( \tau_{nr}/(\tau_r + \tau_{nr}) \)) are consistent with the corresponding values of exfoliated monolayer TMDCs(48).

Combine the Eq. S10 and the definition of conversion efficiency (\( CE \)) in the main text (\( CE = N_{ph0}/N_e \)), \( CE \) can be also expressed as Eq. S11:

\[
CE = \frac{\gamma_e \Gamma_e}{\Gamma_d (\gamma_d - \Gamma_e) + (\gamma_r + \gamma_{nr}) \gamma_d}
\]  (S11)

Eq. S11 allows us to theoretically estimate \( CE \) from the knowledge of the rates of impact excitation (\( \Gamma_e \)), dissociation (\( \Gamma_d \)), exciton recombination (\( \gamma_r \) and \( \gamma_{nr} \)), and leakage of drifting carriers (\( \gamma_d \)). For our devices, these rates can be obtained by substituting the needed parameters listed in Table S3 and Table S4 into Eq. S8, Eq. S9, \( \gamma_d = 2\mu F/W \), \( \gamma_{nr} = 1/\tau_{nr} \) and
\( \gamma_r = 1/\tau_r \). As in the main text, we briefly discuss the values of \( CE \) for different carrier mean free path (\( \lambda_m \)) and exciton binding energy (\( E_b \)), which influence exciton impact excitation rate (\( \Gamma_i \)) and dissociation rate (\( \Gamma_d \)) under a certain electrical field intensity, respectively.

To study the influence of \( \lambda_m \) and \( E_b \) on \( CE \), we take device 6# and 4# as examples, changing \( \lambda_m \) from 4 to 100 nm and \( E_b \) from 20 to 600 meV, respectively, while other parameters of device 6# and 4# are fixed as shown in Table S3 and S4. The \( \lambda_m \) and \( E_b \) dependent \( CE \) are plotted in Fig. 4b and Fig. 4c in the main text, respectively.

Figure 4b (in the main text) clearly indicates that \( CE \) improve with the increase of \( \lambda_m \), due to the increase of impact excitation rate at certain electric field intensity according to Eq. S8. For example, at the electric field intensity around 50 V/\( \mu \)m, as \( \lambda_m \) increases from 4 nm to 14 nm, \( CE \) can achieve an improvement more than three orders. This shows that the short \( \lambda_m \) of TMDC materials severely restricts the values of \( CE \). But in the future, the improvement of material quality will certainly result in a longer \( \lambda_m \) and higher \( CE \) in our proposed device. On the other hand, longer \( \lambda_m \) also allows a lower operating voltage due to the reduction of impact excitation threshold electric field, which is favorable for practical application. Meanwhile, the lower operating voltage will also reduce the exciton dissociation rate.

In addition to \( \lambda_m \), the effect of exciton binding energy (\( E_b \)) on \( CE \) is shown in Fig. 4c (see the main text). The results reveal that the values of \( CE \) increase when \( E_b \) gets larger, this is because larger \( E_b \) greatly suppresses the dissociation (or ionization) of impact-excited excitons, thus allowing more radiative recombination. In conventional semiconductors, due to the small binding energy, the impact excitation mechanism is mostly utilized in sensitive photon detectors. While, in monolayer TMDCs, the extremely large binding energies of 200-600 meV make such monolayers an ideal material choice for such impact excitation EL devices.

Section S7. Comparison between Impact Excitation and Thermal Light Emission

Thermal light emission has been explored in carbon nanotube, graphene, and monolayer MoS\(_2\), which occurs at high electron temperature over 1000 K. Joule heat is generated by the injected current, and the electric fields are usually less than 10 V/\( \mu \)m. To avoid heat dissipation and oxidation of materials, it’s necessary to employ the suspended structures in a vacuum for the devices. While in the impact excitation process, carriers are accelerated by very high electric fields over several tens of V/\( \mu \)m to generate excitons through impact, which don’t need suspended structures and vacuum ambiance.

For a comprehensive description of impact excitation and thermal light emission, the total exciton generation rate \( \Gamma_t \) under a strong electric field can be expressed by the following equation(35):

\[
\Gamma_t = \Gamma_0 \exp \left( -\frac{E_{th}}{\sqrt{\left(k_BT_{op}\right)^2 + (eF\lambda_m)^2}} \right)
\]

(S12)
where $E_{th}$ is impact excitation threshold energy, $\Gamma_0$ is a constant, $T_{op}$ is electron temperature. In this equation, the total exciton generation can be divided into two parts, thermal and impact excitation. For thermal light emission, we assume $T_{op}=1500$ K, then $k_B T_{op}$ is about 129.4 meV. While for impact excitation, we assume $F=100$ V/μm, $\lambda_m=10$ nm, $eF\lambda_m$ is about 1000 meV. Therefore, under a strong electric field, we can conclude that the exciton generation is mainly contributed by the impact excitation process.

Section S8. The Measurement of Carrier Density and Mobility

For estimating the conversion efficiency of our devices, we need to extract the carrier density of our TMDC materials at room temperature. So, we fabricate TMDCs FETs and perform the four-probe measurements, as shown in Fig. S6a. The measurement is divided into two steps. First, we measure the gate voltage ($V_g$) dependent current ($I_{sd}$) and the potential difference between the voltage probes at the same time with a fixed bias voltage ($V_{sd}$), as shown in Fig. S6b. Using the equation for the four-probe device(49) $\mu = (D/WchC_i)/[d((I_{sd}-I_0)/V_{23})/dV_g]$, where $D$ is the distance between the voltage probes, $Wch$ is the channel width, $C_i$ is the capacitance per unit area, and $I_0$ is the current in the subthreshold regime, we can get the carrier mobility $\mu$, as shown in Fig. S6d. Then, as shown in Fig. S6c, we measure the $V_{23}$ dependent $I_{sd}$ without $V_g$ by sweeping $V_{sd}$, and substitute the $\mu$ into the equation $n_0 = (D/e\mu Wch)\times(I_{sd}/V_{23})$, where $e$ is the elementary charge. Employing this method, we fabricate several FETs based on different TMDC materials and obtain the corresponding $n_0$, as shown in Fig. S6e. The values we used to calculate $CE$ are $1 \times 10^{10}$ cm$^{-2}$, $1 \times 10^{12}$ cm$^{-2}$, and $1 \times 10^{10}$ cm$^{-2}$ (as the parameters listed in Table S3) for WSe$_2$, MoSe$_2$, and WS$_2$, respectively.

Figure S6. Pre-existing carrier density and carrier mobility measurements based on four-probe FETs. a, The optical image of a four-probe FET and the measurement schematic. b, The $V_g$ dependent $I_{sd}$ and the voltage probe $V_{23}$. c, The linear relationship between $V_{23}$ and $I_{sd}$. Pre-existing carrier mobilities (d) and carrier densities (e) of TMDC materials in the testing four-probe FETs.
In literature, the reported carrier densities of TMDCs span a wide range due to different growth conditions of monolayer or bulk materials and the introduction of impurities and defects during device fabrication. The methods to measure carrier densities in TMDCs are usually based on the Hall effect, two or four-probe methods, or capacitance model. In our work, we use four-probe method, which is widely used because of its simplicity and good accuracy. The monolayer TMDCs are obtained by mechanical exfoliation from bulk materials purchased from HQ Graphene or 2D Semiconductors. The carrier densities of WS\textsubscript{2} and WSe\textsubscript{2} we measured are in the range of \(10^9\sim10^{11}\ \text{cm}^{-2}\) and \(10^8\sim10^{12}\ \text{cm}^{-2}\), respectively. HQ Graphene's official website lists the carrier densities of their intrinsic WS\textsubscript{2}, WSe\textsubscript{2} at about \(10^{14}\sim10^{15}\ \text{cm}^{-3}\) (equivalent to an areal carrier density of about \(10^{10}\ \text{cm}^{-2}\)). While the carrier densities of MoSe\textsubscript{2} we measured are in the range of \(10^{11}\sim10^{13}\ \text{cm}^{-2}\), which are larger than the values given by the vendors. This may be caused by the introduction of impurities and defects during exfoliation from the bulk materials or device fabrication process, which causes the density level to be comparable to that of MoSe\textsubscript{2} in ref. (50) of about \(6 \times 10^{18}\ \text{cm}^{-3}\) (equivalent to an areal density of about \(10^{12}\ \text{cm}^{-2}\)). Ref (50) also purchased their bulk MoSe\textsubscript{2} from 2D Semiconductors. Overall, we believe that the carrier density levels from our measurement are reasonable.

For carrier mobility, the reported values of monolayer TMDCs are on the order of \(10^1\) to \(10^2\)\(^{51}\). Notice that carrier mobility depends sensitively on annealing conditions and dielectric encapsulation. In our measurement results, the carrier mobility of WSe\textsubscript{2}, WS\textsubscript{2}, and MoSe\textsubscript{2} is mainly distributed in the range of \(10^{-3}\sim10^{-1}\ \text{cm}^{2}\text{V}^{-1}\text{s}^{-1}\). Compared to the reported values in the literature, our measurement results are relatively small, and this may due to the difference of device fabrication process or the doping levels of bulk TMDCs.

**Section S9. Temperature-dependent Light-emitting Intensity**

Figure S7 shows the EL spectra of a monolayer WSe\textsubscript{2} based device under 300 K, 250 K and 200 K. The device operate in a vacuum in a cryostat, excited by 24 V AC square wave with a period of 40 μs. The integration time is 5 seconds. Under the same electrical driving condition, the EL intensities decrease when the temperature drops from 300 K to 200 K. Not only the dark exciton relaxation in WSe\textsubscript{2} limits the EL intensity at low temperature, but also the reduction of the pre-existing free carrier density at low temperature aggravate this trend.

![Figure S7. The temperature-dependent EL spectra of a monolayer WSe\textsubscript{2} based device.](image-url)
REFERENCES AND NOTES

1. K. F. Mak, C. Lee, J. Hone, J. Shan, T. F. Heinz, Atomically ThinMoS$_2$: A new direct-gap semiconductor. *Phys. Rev. Lett.* **105**, 136805 (2010).

2. A. Splendiani, L. Sun, Y. Zhang, T. Li, J. Kim, C.-Y. Chim, G. Galli, F. Wang, Emerging photoluminescence in monolayer MoS$_2$. *Nano Lett.* **10**, 1271–1275 (2010).

3. M. Koperski, K. Nogajewski, A. Arora, V. Cherkez, P. Mallet, J.-Y. Veuillen, J. Marcus, P. Kossacki, M. Potemski, Single photon emitters in exfoliated WSe$_2$ structures. *Nat. Nanotechnol.* **10**, 503–506 (2015).

4. Y.-M. He, G. Clark, J. R. Schaibley, Y. He, M.-C. Chen, Y.-J. Wei, X. Ding, Q. Zhang, W. Yao, X. Xu, C.-Y. Lu, J.-W. Pan, Single quantum emitters in monolayer semiconductors. *Nat. Nanotechnol.* **10**, 497–502 (2015).

5. C. Chakraborty, L. Kinnischtzke, K. M. Goodfellow, R. Beams, A. N. Vamivakas, Voltage-controlled quantum light from an atomically thin semiconductor. *Nat. Nanotechnol.* **10**, 507–511 (2015).

6. A. Srivastava, M. Sidler, A. V. Allain, D. S. Lembke, A. Kis, A. Imamoğlu, Optically active quantum dots in monolayer WS$_2$. *Nat. Nanotechnol.* **10**, 491–496 (2015).

7. B. W. H. Baugher, H. O. H. Churchill, Y. Yang, P. Jarillo-Herrero, Optoelectronic devices based on electrically tunable p-n diodes in a monolayer dichalcogenides. *Nat. Nanotechnol.* **9**, 262–267 (2014).

8. A. Pospischil, M. M. Furchi, T. Mueller, Solar-energy conversion and light emission in an atomic monolayer p-n diode. *Nat. Nanotechnol.* **9**, 257–261 (2014).

9. J. S. Ross, P. Klement, A. M. Jones, N. J. Ghimire, J. Yan, D. G. Mandrus, T. Taniguchi, K. Watanabe, K. Kitamura, W. Yao, D. H. Cobden, X. Xu, Electrically tunable excitonic light-emitting diodes based on monolayer WSe$_2$ p-n junctions. *Nat. Nanotechnol.* **9**, 268–272 (2014).

10. R. S. Sundaram, M. Engel, A. Lombardo, R. Krupke, A. C. Ferrari, Ph. Avouris, M. Steiner, Electroluminescence in single layer MoS$_2$. *Nano Lett.* **13**, 1416–1421 (2013).
11. F. Withers, O. Del Pozo-Zamudio, A. Mishchenko, A. P. Rooney, A. Gholiniia, K. Watanabe, T. Taniguchi, S. J. Haigh, A. K. Geim, A. I. Tartakovskii, K. S. Novoselov, Light-emitting diodes by band-structure engineering in van der Waals heterostructures. *Nat. Mater.* 14, 301–306 (2015).

12. D.-H. Lien, M. Amani, S. B. Desai, G. H. Ahn, K. Han, J.-H. He, J. W. Ager III, M. C. Wu, A. Javey, Large-area and bright pulsed electroluminescence in monolayer semiconductors. *Nat. Commun.* 9, 1229 (2018).

13. F. Withers, O. Del Pozo-Zamudio, S. Schwarz, S. Dufferwiel, P. M. Walker, T. Godde, A. P. Rooney, A. Gholiniia, C. R. Woods, P. Blake, S. J. Haigh, K. Watanabe, T. Taniguchi, I. L. Aleiner, A. K. Geim, V. I. Fal’ko, A. I. Tartakovskii, K. S. Novoselov, WSe$_2$ light-emitting tunneling transistors with enhanced brightness at room temperature. *Nano Lett.* 15, 8223–8228 (2015).

14. S. Wang, J. Wang, W. Zhao, F. Giustiniano, L. Chu, I. Verzhbitskiy, J. Zhou Yong, G. Eda, Efficient carrier-to-exciton conversion in field emission tunnel diodes based on MIS-type van der Waals heterostack. *Nano Lett.* 17, 5156–5162 (2017).

15. Y.-Q. Bie, G. Grosso, M. Heuck, M. M. Furchi, Y. Cao, J. Zheng, D. Bunandar, E. Navarro-Moratalla, L. Zhou, D. K. Efetov, T. Taniguchi, K. Watanabe, J. Kong, D. Englund, P. Jarillo-Herrero, A MoTe$_2$-based light-emitting diode and photodetector for silicon photonic integrated circuits. *Nat. Nanotechnol.* 12, 1124–1129 (2017).

16. Y. J. Zhang, T. Oka, R. Suzuki, J. T. Ye, Y. Iwasa, Electrically switchable chiral light-emitting transistor. *Science* 344, 725–728 (2014).

17. Y. Zhu, Z. Li, L. Zhang, B. Wang, Z. Luo, J. Long, J. Yang, L. Fu, Y. Lu, High-efficiency monolayer molybdenum ditelluride light-emitting diode and photodetector. *ACS Appl. Mater. Interfaces* 10, 43291–43298 (2018).

18. J. Cho, M. Amani, D.-H. Lien, H. Kim, M. Yeh, V. Wang, C. Tan, A. Javey, Centimeter-scale and visible wavelength monolayer light-emitting devices. *Adv. Funct. Mater.* 30, 1907941 (2020).
19. S. Wu, S. Buckley, J. R. Schaibley, L. Feng, J. Yan, D. G. Mandrus, F. Hatami, W. Yao, J. Vučković, A. Majumdar, X. Xu, Monolayer semiconductor nanocavity lasers with ultralow thresholds. *Nature* **520**, 69–72 (2015).

20. Y. Ye, Z. J. Wong, X. Lu, X. Ni, H. Zhu, X. Chen, Y. Wang, X. Zhang, Monolayer excitonic laser. *Nat. Photonics* **9**, 733–737 (2015).

21. Y. Li, J. Zhang, D. Huang, H. Sun, F. Fan, J. Feng, Z. Wang, C. Z. Ning, Room-temperature continuous-wave lasing from monolayer molybdenum ditelluride integrated with a silicon nanobeam cavity. *Nat. Nanotechnol.* **12**, 987–992 (2017).

22. E. Y. Paik, L. Zhang, G. W. Burg, R. Gogna, E. Tutuc, H. Deng, Interlayer exciton laser of extended spatial coherence in atomically thin heterostructures. *Nature* **576**, 80–84 (2019).

23. Y. Liu, H. Fang, A. Rasmita, Y. Zhou, J. Li, T. Yu, Q. Xiong, N. Zheludev, J. Liu, W. Gao, Room temperature nanocavity laser with interlayer excitons in 2D heterostructures. *Sci. Adv.* **5**, eaav4506 (2019).

24. C.-Z. Ning, Semiconductor nanolasers and the size-energy-efficiency challenge: A review. *Adv. Photonics* **1**, 014002 (2019).

25. Z. Wang, H. Sun, Q. Zhang, J. Feng, J. Zhang, Y. Li, C.-Z. Ning, Excitonic complexes and optical gain in two-dimensional molybdenum ditelluride well below the Mott transition. *Light Sci. Appl.* **9**, 39 (2020).

26. H. Fang, J. Liu, H. Li, L. Zhou, L. Liu, J. Li, X. Wang, T. F. Krauss, Y. Wang, 1305 nm few-layer MoTe$_2$-on-silicon laser-like emission. *Laser Photonics Rev.* **12**, 1800015 (2018).

27. M. H. D. Guimarães, H. Gao, Y. Han, K. Kang, S. Xie, C.-J. Kim, D. A. Muller, D. C. Ralph, J. Park, Atomically thin ohmic edge contacts between two-dimensional materials. *ACS Nano* **10**, 6392–6399 (2016).

28. X. Cui, E.-M. Shih, L. A. Jauregui, S. H. Chae, Y. D. Kim, B. Li, D. Seo, K. Pistunova, J. Yin, J.-H. Park, H.-J. Choi, Y. H. Lee, K. Watanabe, T. Taniguchi, P. Kim, C. R. Dean, J. C. Hone, Low-
temperature ohmic contact to monolayer MoS$_2$ by van der Waals bonded Co/h-BN electrodes. *Nano Lett.* **17**, 4781–4786 (2017).

29. S. Cho, S. Kim, J. H. Kim, J. Zhao, J. Seok, D. H. Keum, J. Baik, D.-H. Choe, K. J. Chang, K. Suenaga, S. W. Kim, Y. H. Lee, H. Yang, Phase patterning for ohmic homojunction contact in MoTe$_2$. *Science* **349**, 625–628 (2015).

30. A. Allain, J. Kang, K. Banerjee, A. Kis, Electrical contacts to two-dimensional semiconductors. *Nat. Mater.* **14**, 1195–1205 (2015).

31. H. Li, G. Lu, Y. Wang, Z. Yin, C. Cong, Q. He, L. Wang, F. Ding, T. Yu, H. Zhang, Mechanical exfoliation and characterization of single- and few-layer nanosheets of WSe$_2$, TaS$_2$, and TaSe$_2$. *Small* **9**, 1974–1981 (2013).

32. Y. Zhan, Z. Liu, S. Najmaei, P. M. Ajayan, J. Lou, Large-area vapor-phase growth and characterization of MoS$_2$ atomic layers on a SiO$_2$ substrate. *Small* **8**, 966–971 (2012).

33. A. Castellanos-Gomez, M. Buscema, R. Molenaar, V. Singh, L. Janssen, H. S. J. van der Zant, G. A. Steele, Deterministic transfer of two-dimensional materials by all-dry viscoelastic stamping. *2D Mater.* **1**, 011002 (2014).

34. J. Chen, V. Perebeinos, M. Freitag, J. Tsang, Q. Fu, J. Liu, P. Avouris, Bright infrared emission from electrically induced excitons in carbon nanotubes. *Science* **310**, 1171–1174 (2005).

35. V. Perebeinos, P. Avouris, Impact excitation by hot carriers in carbon nanotubes. *Phys. Rev. B* **74**, 121410 (2006).

36. Y. Okuto, C. R. Crowell, Energy-conservation considerations in the characterization of impact ionization in semiconductors. *Phys. Rev. B* **6**, 3076–3081 (1972).

37. L. Dobusch, S. Schuler, V. Perebeinos, T. Mueller, Thermal light emission from monolayer MoS$_2$. *Adv. Mater.* **29**, 1701304 (2017).
38. I. Yang, Z. Li, J. Wong-Leung, Y. Zhu, Z. Li, N. Gagrani, L. Li, M. N. Lockrey, H. Nguyen, Y. Lu, H. H. Tan, C. Jagadish, L. Fu, Multiwavelength single nanowire InGaAs/InP quantum well light-emitting diodes. *Nano Lett.* **19**, 3821–3829 (2019).

39. V. Wood, J. E. Halpert, M. J. Panzer, M. G. Bawendi, V. Bulović, Alternating current driven electroluminescence from ZnSe/ZnS:Mn/ZnS nanocrystals. *Nano Lett.* **9**, 2367–2371 (2009).

40. J. Benoit, C. Barthou, P. Benalloul, K. Polamo, The electro-optical behavior of SrS:Ce electroluminescent devices under photonic excitation. *J. Appl. Phys.* **87**, 198–202 (2000).

41. D. Bozyigit, V. Wood, Y. Shirasaki, V. Bulovic, Study of field driven electroluminescence in colloidal quantum dot solids. *J. Appl. Phys.* **111**, 113701 (2012).

42. L. Wang, L. Xiao, H. Gu, H. Sun, Advances in alternating current electroluminescent devices. *Adv. Opt. Mater.* **7**, 1801154 (2019).

43. F. Fan, S. Turkdogan, Z. Liu, D. Shelhammer, C. Z. Ning, A monolithic white laser. *Nat. Nanotechnol.* **10**, 796–803 (2015).

44. I. Fushman, D. Englund, J. Vučković, Coupling of PbS quantum dots to photonic crystal cavities at room temperature. *Appl. Phys. Lett.* **87**, 241102 (2005).

45. J. D. Jackson, *Classical Electrodynamics Third Edition* (Wiley, ed. 3, 1998).

46. M. Massicotte, F. Vialla, P. Schmidt, M. B. Lundeberg, S. Latini, S. Haastrup, M. Danovich, D. Davydovskaya, K. Watanabe, T. Taniguchi, V. I. Fal’ko, K. S. Thygesen, T. G. Pedersen, F. H. L. Koppens, Dissociation of two-dimensional excitons in monolayer WSe$_2$. *Nat. Commun.* **9**, 1633 (2018).

47. T. Brumme, M. Calandra, F. Mauri, Determination of scattering time and of valley occupation in transition-metal dichalcogenides doped by field effect. *Phys. Rev. B* **93**, 081407 (2016).
48. M. Amani, P. Taheri, R. Addou, G. H. Ahn, D. Kiriya, D.-H. Lien, J. W. Ager, R. M. Wallace, A. Javey, Recombination kinetics and effects of superacid treatment in sulfur- and selenium-based transition metal dichalcogenides. *Nano Lett.* **16**, 2786–2791 (2016).

49. V. Podzorov, M. E. Gershenson, Ch. Kloc, R. Zeis, E. Bucher, High-mobility field-effect transistors based on transition metal dichalcogenides. *Appl. Phys. Lett.* **84**, 3301–3303 (2004).

50. H. I. Jeong, S. Park, H. I. Yang, W. Choi, Electrical properties of MoSe$_2$ metal-oxide-semiconductor capacitors. *Mater. Lett.* **253**, 209–212 (2019).

51. S.-L. Li, K. Tsukagoshi, E. Orgiu, P. Samori, Charge transport and mobility engineering in two-dimensional transition metal chalcogenide semiconductors. *Chem. Soc. Rev.* **45**, 118–151 (2015).