Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

AC-Driven Multicolor Electroluminescence from a Hybrid WSe_2 Monolayer/AlGaInP Quantum Wells Light-Emitting Device

Ya-Hui Chang[1,2], *Yen-Shou Lin*[1,2], *Konthoujam James Singh*[2], *Hsiang-Ting Lin*[1], *Chiao-Yun Chang*[1,3], *Zheng-Zhe Chen*[1,4], *Yu-Wei Zhang*[1,5], *Shih-Yen Lin*[1,5], *Hao-Chung Kuo*[1,2], *and Min-Hsiung Shih*[1,2,6*]

[1]Research Center for Applied Sciences (RCAS), Academia Sinica, Taipei 11529, Taiwan

[2]Department of Photonics and Institute of Electro-Optical Engineering, College of Electrical and Computer Engineering, National Yang Ming Chiao Tung University, Hsinchu 30010, Taiwan

[3]Department of Electrical Engineering, National Taiwan Ocean University, Keelung 202301, Taiwan

[4]Department of Physics, National Taiwan University, Taipei, Taiwan, Taipei 10617, Taiwan

[5]Graduate Institute of Electronics Engineering, National Taiwan University, Taipei 10617, Taiwan

[6]Department of Photonics, National Sun Yat-sen University, Kaohsiung 80424, Taiwan

*E-mail address: mhshih@gate.sinica.edu.tw

1) Material Preparation and Device Fabrication

The CVD-grown WSe_2 monolayer on a sapphire substrate was transferred onto a red AlGaInP-based LED by using the standard poly(methyl methacrylate) (PMMA)-assisted transfer method. In this method, the PMMA solution was spin-coated onto the WSe_2 to act as a supporting layer. The PMMA-coated sample was placed in buffered oxide etch (BOE) to etch the sapphire and then peel the PMMA–WSe_2 film from the sapphire substrate. The obtained PMMA–WSe_2 film was rinsed with deionized (DI) water to reduce the amount of residual BOE. Before the PMMA–WSe_2 floating on the surface of the DI water was pick-up by the target substrate of the AlGaInP-based LED, AlO_x film was deposited on the AlGaInP-based LED substrate through atomic layer deposition, which was performed using trimethylaluminum and water vapor as the precursors at 180 °C and a growth rate of 1.0 Å/cycle. An AlO_x film of thickness 30 nm was deposited as a spacer layer between the WSe_2 monolayer and AlGaInP-based LED wafer, and the part of the AlO_x film was then selectively removed to create the gate electrode. Next, the as-transferred WSe_2 was soaked in acetone to dissolve the PMMA layer, and this was followed by selective plasma etching with inductively coupled plasma by using the developed resist employed as the etch mask. This led to the electroluminescence of the WSe_2 monolayer occurring on a limited region of the WSe_2. Finally, through electron-beam evaporation, electrodes consisting of 20 nm titanium as the adhesion layer and 200 nm silver were deposited on the top of the WSe_2 and exposed AlGaInP-based LED.

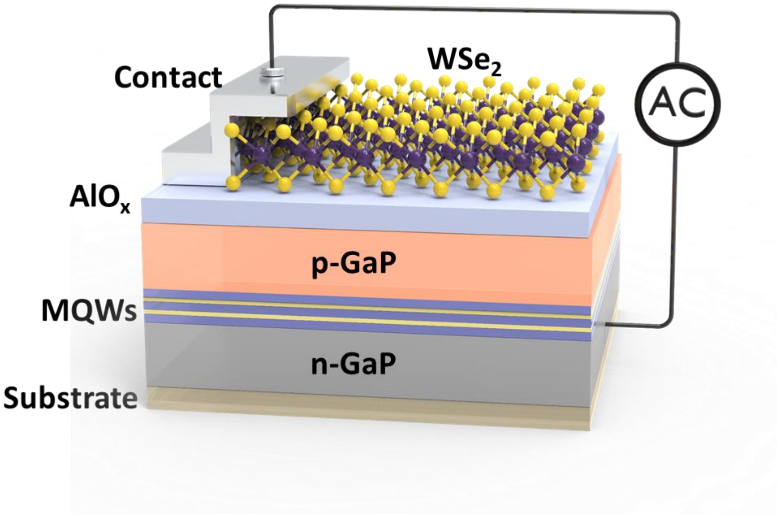

Fig. S1. Device architecture.

2) Estimation of External Quantum Efficiency (EQE) for WSe_2 Monolayer and AlGaInP QWs

The internal quantum efficiency (IQE) can be estimated by $IQE(\%) = \frac{\Phi}{G} \times 100\%$. (*Nano Lett.* 14, 4125–4130(2014))

Here, the generation rate G (s^{-1}) is calculated by the formula,

$$\text{Generation Rate } (s^{-1}) = \frac{P \cdot \alpha \cdot (1-R)}{h\nu},$$

P is the excitation power, α is the absorption coefficient, R is the reflectance and hν is the energy of the 450 nm laser. And $\Phi = \eta \int \frac{I_{PL}}{t_I} d\lambda$ is the photon generated per second. η is the collected photon number coefficient in this measurement system, t_I is the integration time of PL measurement and I_{PL} is the PL intensity. The generation rate can also be presented in (cm^{-2} s^{-1}) unit, which considers the generation rate per area (pumping spot size). The diameter of laser spot is approximately 1.2 μm.

The external quantum efficiency can be estimated using the equation

$$\eta_e = \eta_{I\ ext}$$

, where η_I is the internal quantum efficiency and η_{ext} is the light extraction efficiency. The generation rate per square centimeter of the device is approximately 10^{16}-10^{21} $cm^{-2}s^{-1}$, and the measured internal quantum efficiency, η_I of the WSe_2 monolayer increase approximately 0.08-0.91 % in the carrier concentration range. The extraction efficiency is calculated by using $(4n^2)^{-1}$, and n is the effective refractive index of the medium including the WSe_2 monolayer and the Al_2O_3 layer. The LED device was operated under AC conditions with a voltage of 12 volt and a frequency of 1 MHz at room temperature. Using this formula, the maximum EQE value, the estimated η_e is approximately 0.018 %. The IQE and EQE values are consistent with the reported values in references, such as *Nat. Comm.* 9:1229 (2018), and *Nano Lett.* 22, 5316(2022). The EQE of the AlGaInP QWs is approximately 0.056% with the same estimation method.

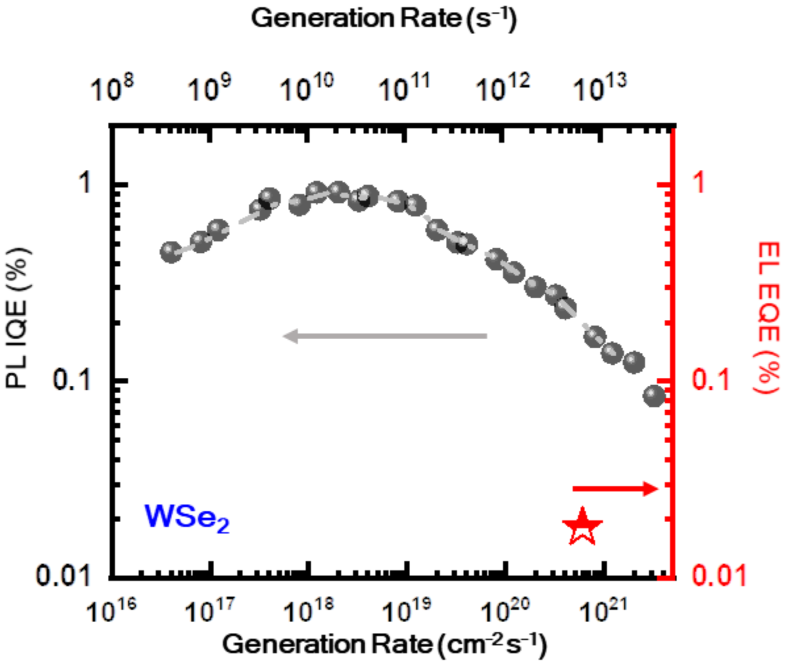

Fig. S2 The measured quantum efficiency of the WSe_2 monolayer LED device.

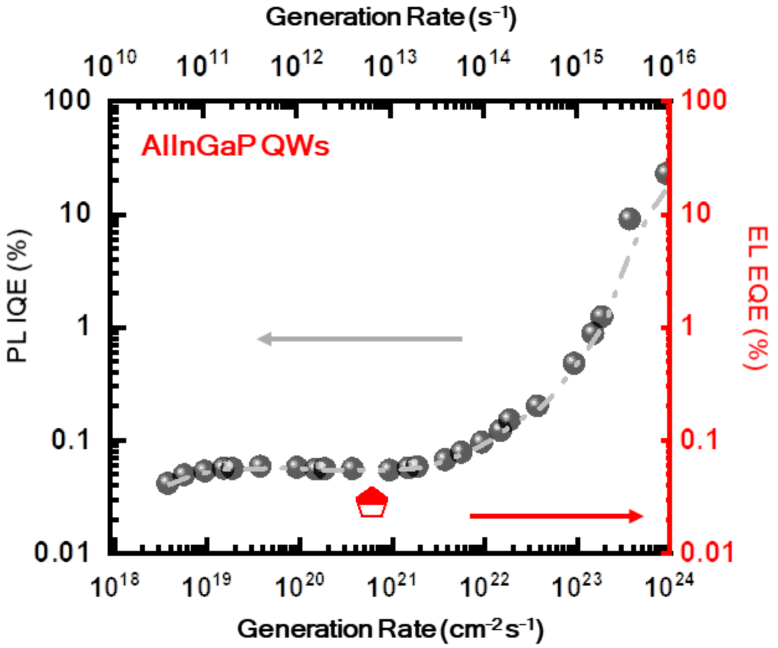

Fig. S3 The measured quantum efficiency of the AlGaInP QWs LED device.