Supplementary Information

Self-Powered Broadband Photodetection Enabled by the Facile CVD-Grown MoS_2/GaN Heterostructures

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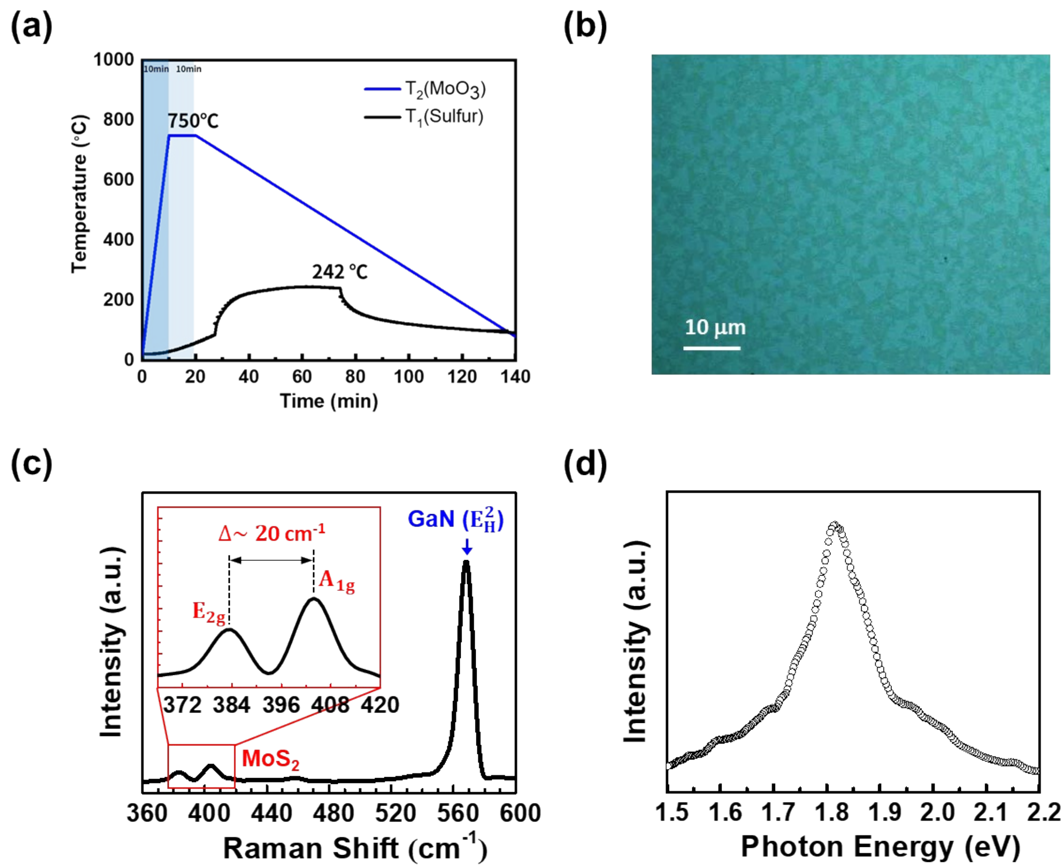

Figure S1. (a) The temperature as a function of time recorded at two regions during the CVD synthesis process. Black and blue curves indicate S and MoO_3 sources, respectively. **(b)** The optical microscope image of CVD growth monolayer MoS_2 on GaN substrate. **(c)** Raman spectrum demonstrating the both spectroscopic analysis of MoS_2 and GaN. **(d)** PL spectrum presenting the optical bandgap of ~1.82 eV.

Figure S1a displays the temporal relationship between temperature and time, as observed in two distinct heating regions, namely T_1 and T_2, during the CVD synthesis process. The temperature profiles for S (black curve) and MoO_3 (blue curve) are depicted. Initially, the temperature in region 1 (S) was set to 200℃, while in region 2 (MoO_3), it was set to 750℃. Subsequently, once the desired temperature was attained, both regions commenced their cooling phase. **Figure S1b** demonstrates the optical microscope (OM) image of the monolayer MoS_2 on GaN using the CVD method. **Figure S1c** presents the Raman spectrum of MoS_2 and GaN, in which the E^1_{2g} mode at ~383 cm^{-1} corresponding to the in-plane vibrational mode, while the A^1_g mode at ~403 cm^{-1} corresponding to the out-of-plane vibrational mode. The separation between these two peaks of ~20 cm^{-1} indicates the monolayer MoS_2. Additionally, the peak appearing at ~568 cm^{-1} was consistent with the E_H^2 p mode of GaN grown on sapphire substrates.[2] **Figure S1d** exhibits the PL spectrum presenting the optical bandgap of ~1.82 eV. These findings are in agreement with previously report.[3]

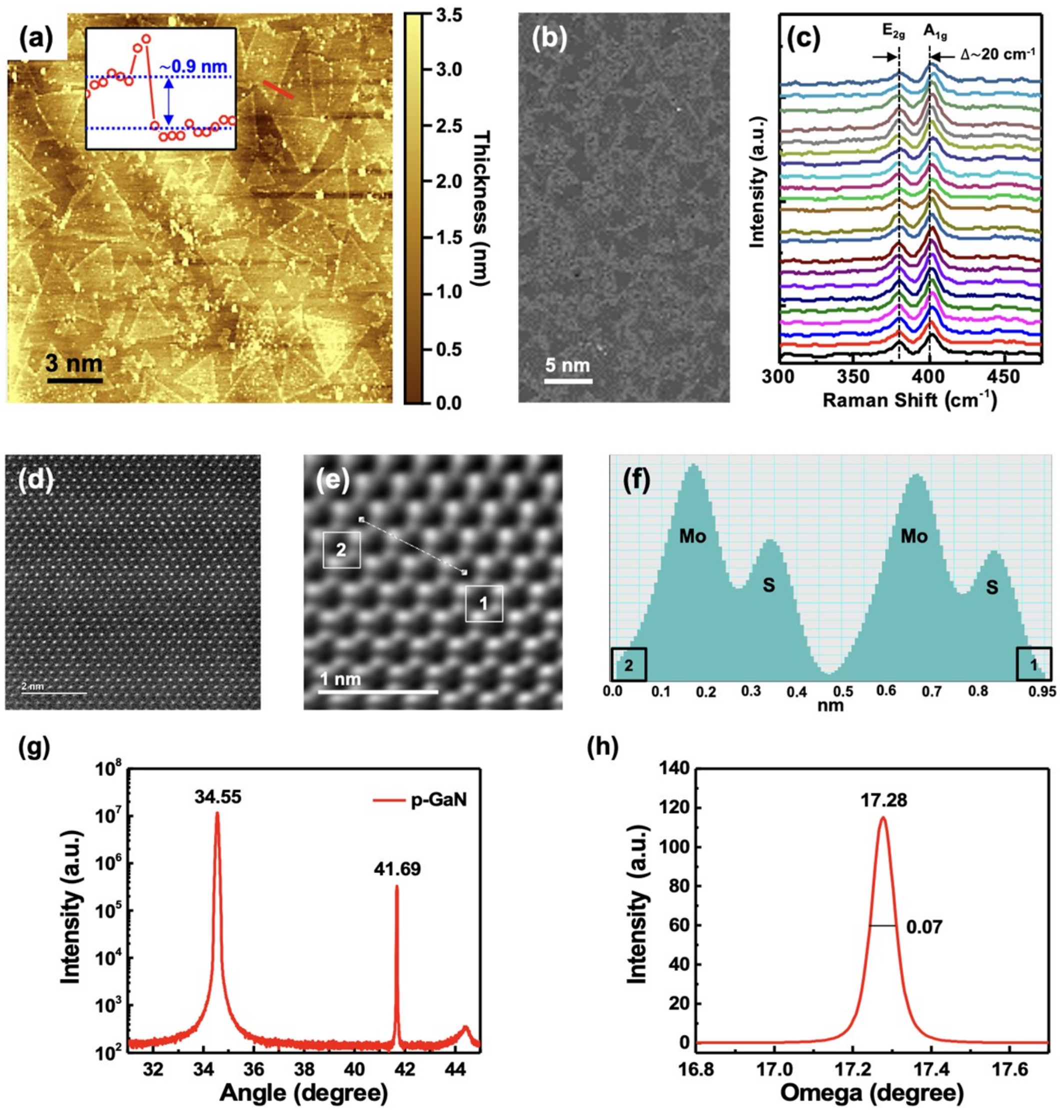

Figure S2. (a) AFM image of MoS_2 on GaN substrate. The inset presents the thickness of MoS_2 is ~0.9 nm. **(b)** SEM image of MoS_2 on GaN substrate. **(c)** Multiple Raman spectra measured indivisually on 22 pieces of MoS_2 in the region of Figure (b), demonstrating the uniformity of MoS_2 layer with Δ~20 cm^{-1}. **(d-e)** Large and small scale of high-resolusion TEM images of MoS_2, respectively. **(f)** The distribution of Mo and S atoms along the white dashed line indicated in Figure (e). **(g-h)** The XRD of p-type GaN substrate.

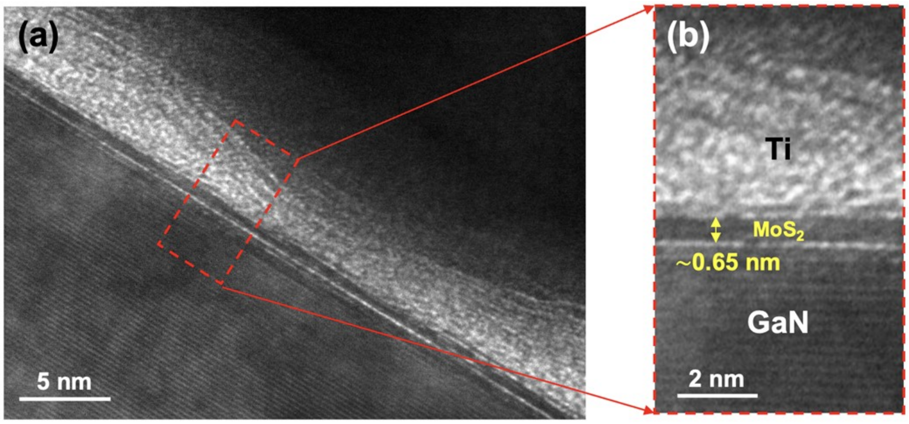

Figure S3. (a) High-resolution TEM image showing a clean interface between monolayer MoS_2 and GaN substrate. **(b)** An enlarged part (red dashed line) of the TEM image from Figure (a) demonstrating monolayer MoS_2 attached on GaN substrate smoothly.

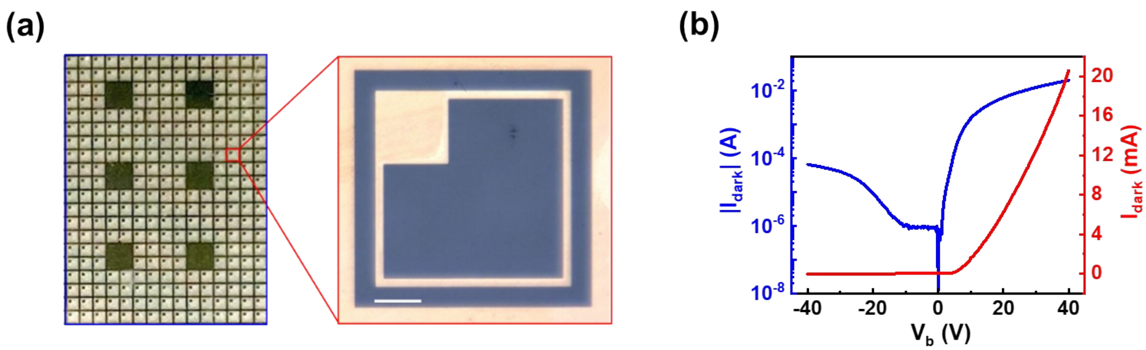

Figure S4. (a) Optical microscope images of the MoS_2/GaN photodetector array. The enlarged image shows a single unit of the photodetector. The scale bar is 50 μm. **(b)** The logarithm (blue curve) and linear (red curve) dark current of the MoS_2/GaN photodetector.

Figure S4a shows the OM images of the MoS_2/GaN photodetector array, in which the enlarged image presents a single unit of the photodetector. **Figure S4b** demonstrates the logarithm (blue curve) and linear (red curve) dark current of the MoS_2/GaN photodetector with a coverage of MoS_2 flakes of ~52.2%. The observed diode-like rectifying behavior confirms the existence of an internal electrical field at the MoS_2/GaN heterojunction. As MoS_2 is known to be an intrinsic n-type semiconductor, the resulting n-MoS_2/p-GaN heterojunction can be compared to a P-N junction. Therefore, the PN junction diode model can be utilized to estimate the ideal factor in the linear region of the IV curve (ranging from 1 V to 8 V):

$$I = I_S[\exp\left(\frac{eV}{\eta KT}\right) - 1] \tag{S1}$$

where I_S of ~0.9 μA is the saturation current at negative bias voltages (ranging from 0 V to -10 V); η is the ideality factor; e, K, and T are the electron charge, Boltzmann's constant and absolute temperature, respectively.[4] By applying a positive bias voltage ranging from 0 V to 10 V, an approximate ideal factor of ~40 can be inferred. The substantial deviation in the ideality factor, which is significantly higher than expected, can be attributed to the presence of interface states between MoS_2 and GaN.[5] These interface states introduce considerable deviations from the anticipated behavior of thermionic emission current and consequently lead to the involvement of alternative transport mechanisms, such as tunneling. This deviation ultimately results in an increased ideality factor for the P-N junction photodetector.

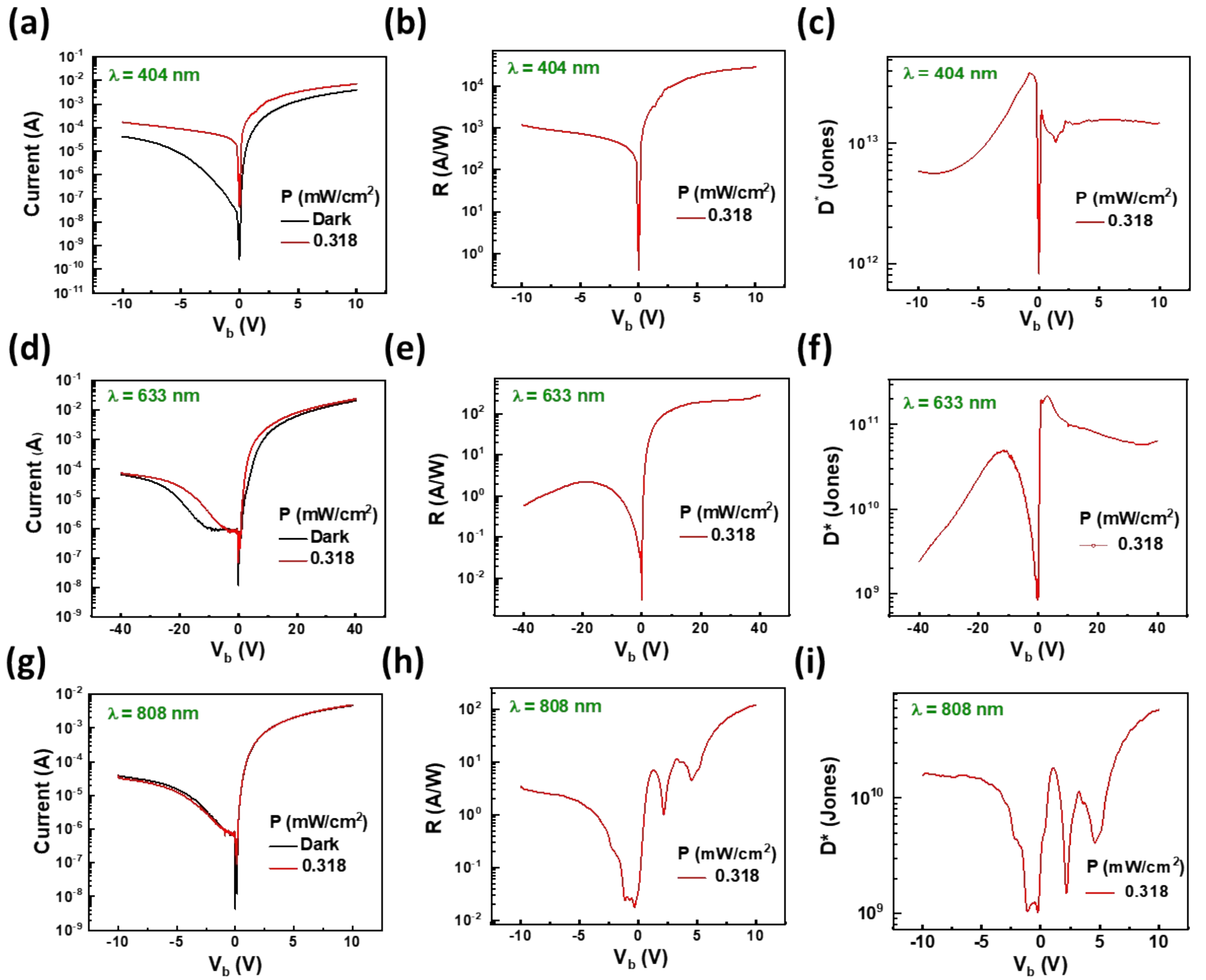

Figure S5. Dark current (black curve), photoresponse, responsivity (R) and specific detectivity (D^*) characteristics of the photodetector illuminated with **(a-c)** 404 nm, **(d-f)** 633 nm and **(g-i)** 808 nm laser with the power density of ~0.318 mW/cm^2, respectively.

Figure S5 exhibits the characteristics of the photodetector, including the dark current (depicted by the black curve), photoresponse, responsivity (R), and specific detectivity (D^*), under different illumination conditions. The device was illuminated with lasers at wavelengths of 404 nm (**Fig. S5a-S5c**), 633 nm (**Fig. S5d-S5f)**, and 808 nm (**Fig. S5g-S5i)**, with a power density of ~0.318 mW/cm^2. These results highlight the remarkable optical detection capabilities of the photodetector across a wide range of wavelengths, encompassing visible light, ultraviolet A (UVA), and near-infrared (NIR) regions.

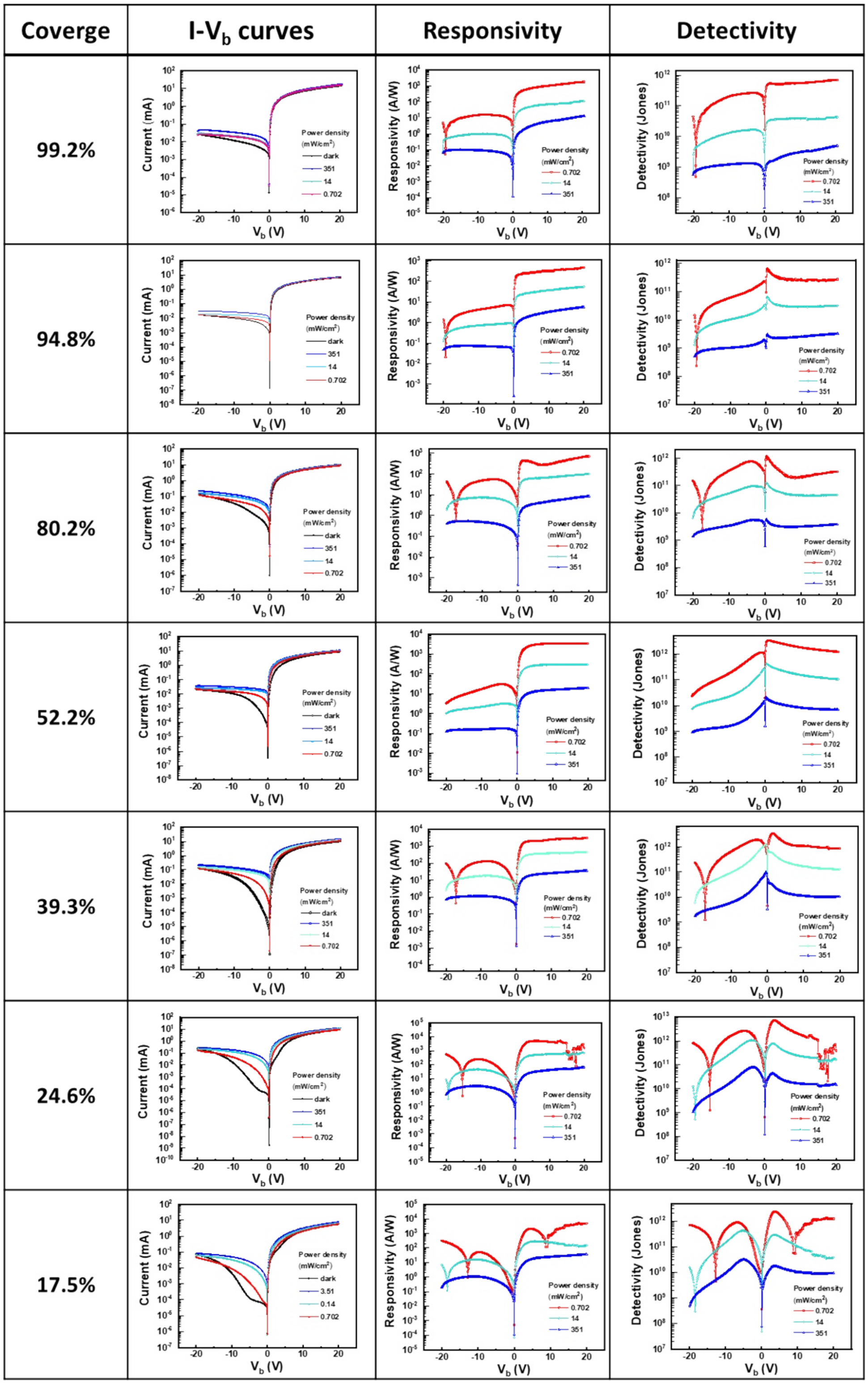

Figure S6. The characteristics of I-V curves, R and D^* with various laser power density for different coverage of MoS_2.

Figure S6 illustrates the I-V characteristics of MoS_2/GaN photodetectors with varying MoS_2 coverage. The photodetectors were assessed under 532 nm laser irradiation, with power densities of approximately 0.7, 14, and 351 mW/cm^2, while bias voltages ranged from -20 V to 20 V. Notably, the photodetectors demonstrate a substantial photoresponse upon illumination, which exhibits an upward trend as the incident light power density increases. Interestingly, the responsivity (R) and specific detectivity (D^*) of the photodetector decrease as the MoS_2 coverage increases. Conversely, they increase when the incident light power density decreases.

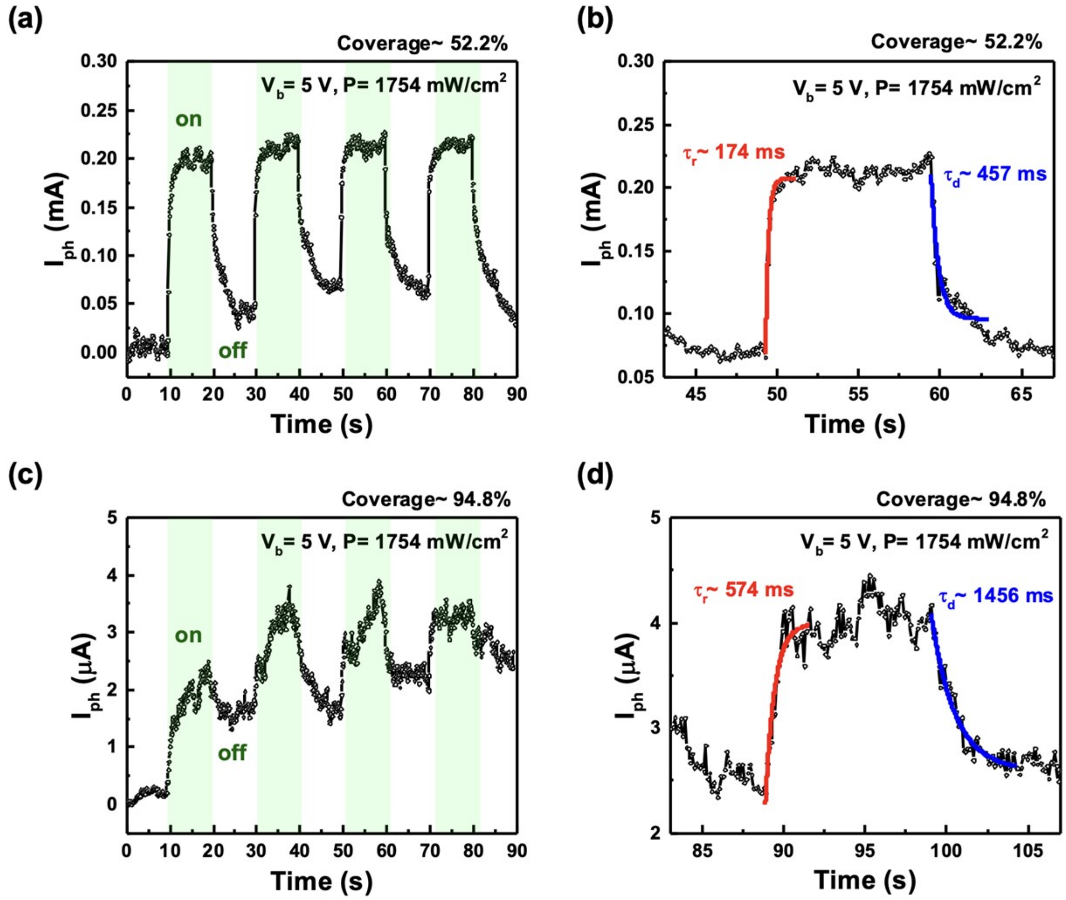

Figure S7. Time-resolved photoresponse of the photodetector illuminated with 532 nm laser under the conditions of $V_b = 5$ V, P = 1754 mW/cm^2 for the coverage percentage of **(a)** ~52.2% and **(b)** ~94.8%, respectively. **(b, d)** The corresponding single period of the photoresponse showing the rise time τ_r (red curves) and decay time τ_d (blue curves) of the photodetector under the two different coverage percentages.

References

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