Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Ultrafast-temporal-responsive flexible photodetector with high sensitivity based on high-crystallinity 2D organic-inorganic perovskite

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Derivation

Derivation of the relationship between responsivity (R_λ) and surface-recombination-velocity (s) is conducted. At first, the equation of responsivity and optical gain is cited from a previous report:[1]

$$R_\lambda = \frac{\eta(\lambda)G(\lambda)}{1}\frac{e\lambda}{hc}, \qquad (1)$$

where λ is excited light wavelength, $\eta(\lambda)$ is efficiency of light absorption, $G(\lambda)$ is optical gain, e is electronic charge, h is planck constant, and c is velocity of light. The optical gain can be written as:[2]

$$G(\lambda) = \frac{\tau_{eff}}{\tau_t}, \qquad (2)$$

where τ_{eff} is effective lifetime of carriers, and τ_t is transit time, expressed as: $1/\tau_t=\mu V/L^2$.

For 2D structure, the τ_{eff} can be expressed as:[3]

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau_b} + \frac{2s}{L}, \qquad (3)$$

where τ_b is bulk carrier lifetime, and L is length of the material. Substituting Equations (2) and (3) into (1), the responsivity can be written as:

$$R_\lambda = \frac{1}{\frac{1}{\mu\tau_b} + \frac{2s}{\mu L}} \cdot \frac{e\eta\lambda V}{hcL^2}, \qquad (4)$$

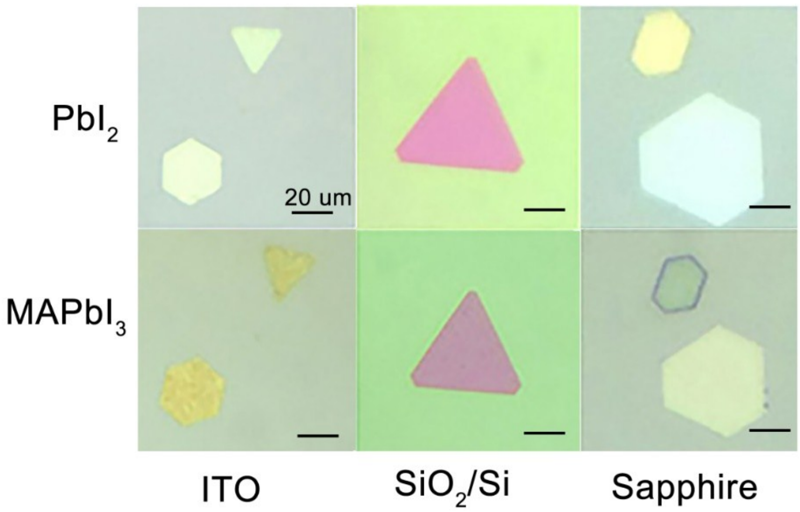

Figure S1. Optical images of 2D $MAPbI_3$ and PbI_2 precursors on ITO, SiO_2/Si and sapphire substrates, indicating low restriction of the substrates.

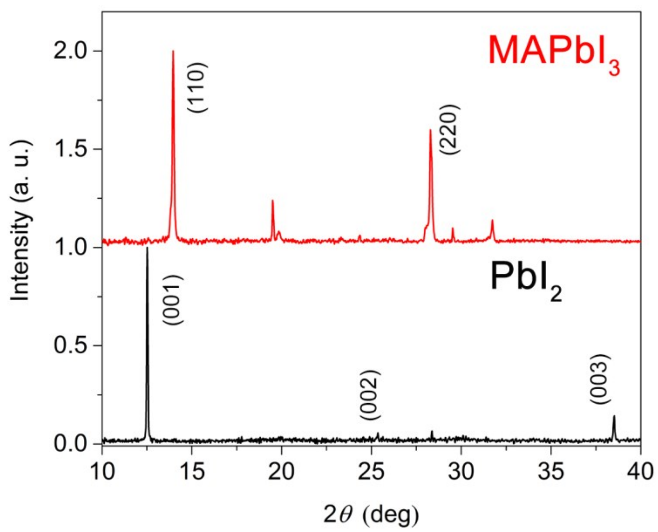

Figure S2. XRD patterns of 2D $MAPbI_3$ (red line) and PbI_2 precursor (black line). The differences of the XRD spectra between 2D layered PbI_2 precursor and $MAPbI_3$ indicate that hexagonal layered PbI_2 precursor has transformed to tetragonal $MAPbI_3$structure.

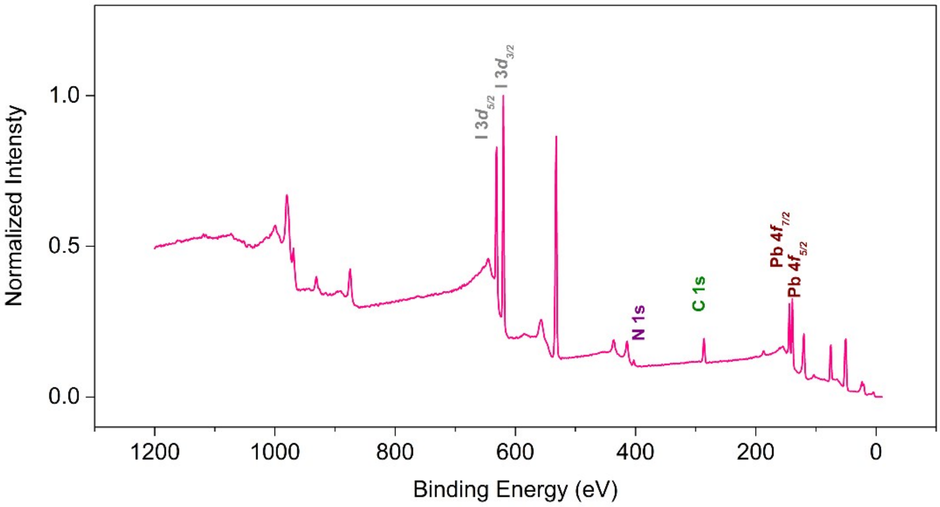

Figure S3. XPS of as-grown 2D $MAPbI_3$. The characteristic peaks of Pb 4*f*, C 1*s*, N 1*s* and I 3*d* are pointed out, which are discussed in the manuscript.

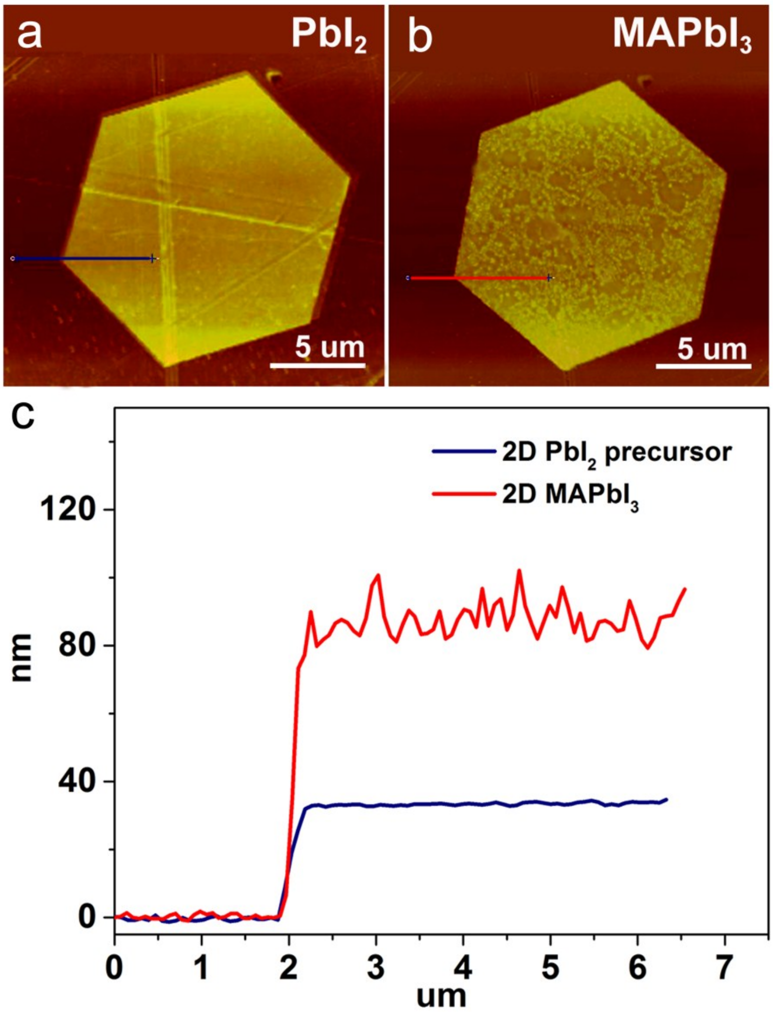

Figure S4. (a) and (b) show AFM images of the 2D layered PbI_2 precursor and corresponding 2D $MAPbI_3$ crystal after gas reaction, respectively. (c) Thicknesses of the samples shown in (a) and (b) exhibit obvious variation.

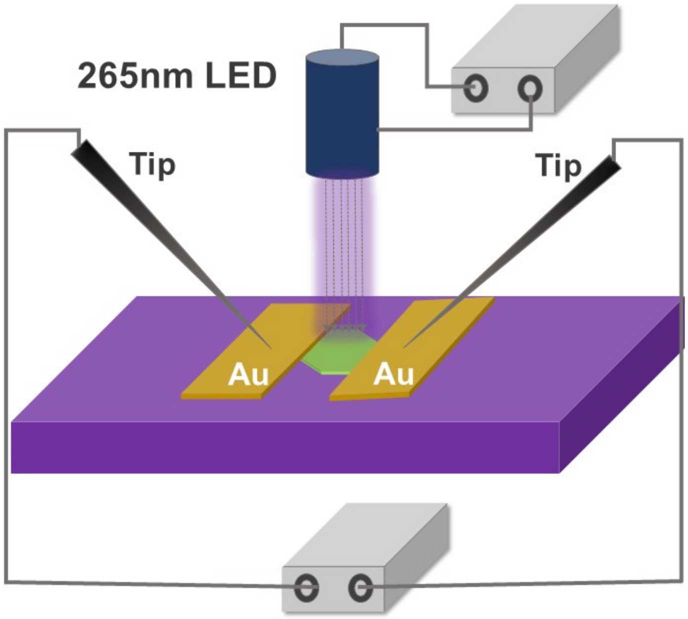

Figure S5. A typical schematic diagram of the electric measurement system, including two tips contacting Au electrodes, a sourcemeter to measure I-V curve and a source to modulate the 265 nm LED light.

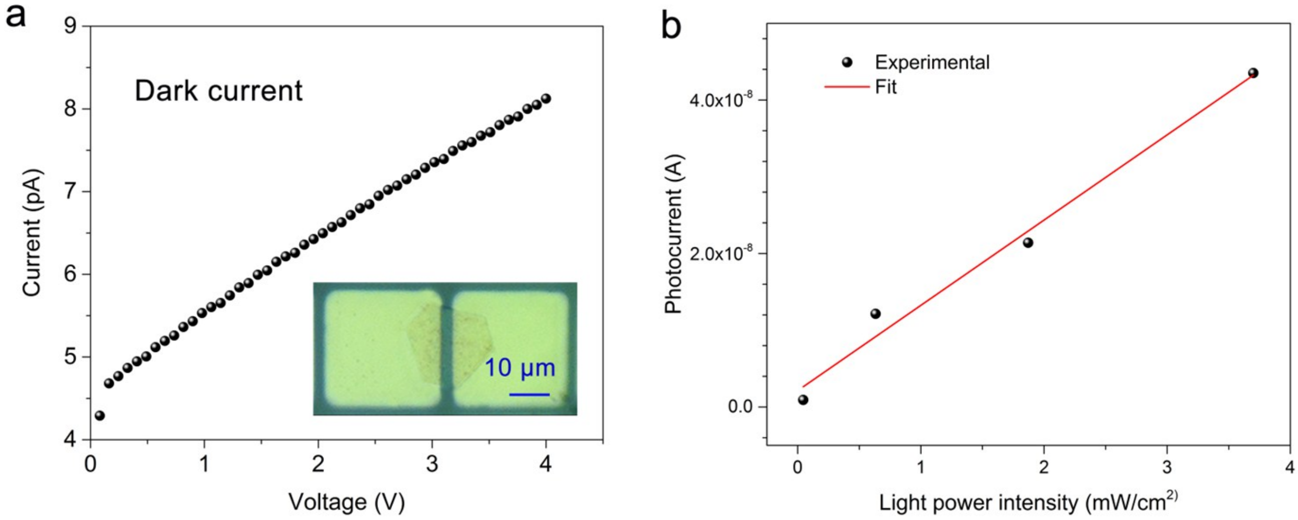

Figure S6. (a) Dark current of the present detector. The inset is corresponding optical photo. (b) Photocurrent as a function of illumination intensity shows a linear increase with increasing illumination intensity, demonstrating a typical photoconductive effect.

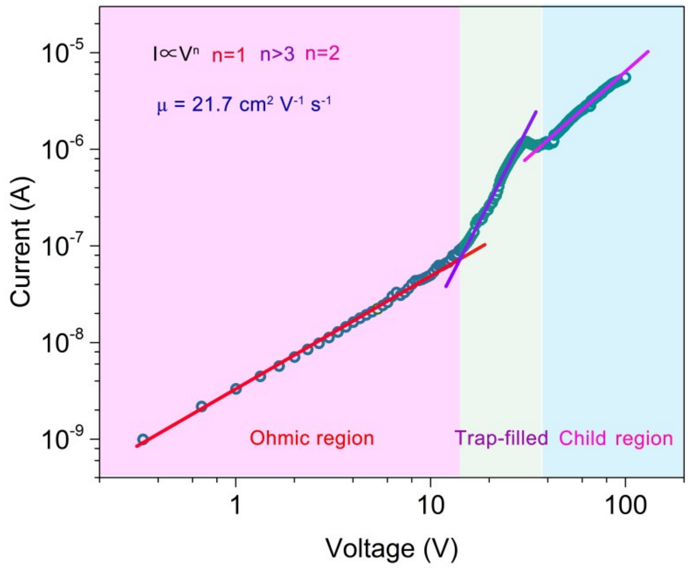

Figure S7. I-V characteristic of bulk $MAPbI_3$ single crystal with a size of 2mm(length)×2mm(width) ×0.5mm(thickness) synthesized via typical inverse temperature crystallization. The I-V characteristic analyzed by space charge limit current effect shows an ohmic region, a trap-filled region and a Child region. The carrier mobility of $MAPbI_3$ can be fitted via the equation: $I=9\varepsilon\varepsilon_0\mu V^2/8L$, where ε is dielectric constant of the material, ε_0 is vacuum dielectric constant, L is length of the material, and the extracted μ=21.7 $cm^2V^{-1}s^{-1}$.

References

1. Saran, R. & Curry, R. J. Lead sulphide nanocrystal photodetector technologies. *Nat. Photon.* **10**, 81-92 (2016).
2. Neamen, D. A. Semiconductor Physics and Devices: Basic Principles. McGraw-Hill, Beijing (2003).
3. Demichel, O. *et al.* Surface Recombination Velocity Measurements of Efficiently Passivated Gold-Catalyzed Silicon Nanowires by a New Optical Method. *Nano Lett.* **10**, 2323-2329 (2010).