Supporting Information

Ultralow dark current and high specific detectivity of Ga_2O_3-based solar-blind photodetector arrays realized via post-annealing in oxygen plasma

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Supporting Note 1: Detailed description of a remote plasma enhanced chemical vapor deposition system

After obtaining Ga_2O_3 thin film, we subjected it to a series of treatments under different conditions using a remote plasma enhanced chemical vapor deposition system. **Figure S1** shows the schematic diagram (a) and photograph (b) of the remote plasma enhanced chemical vapor deposition system. This system mainly consists of several modules: a temperature-controllable tubular furnace, a vacuum pump, a quartz tube, a distance controller, a plasma generator, and an intelligent control system. The plasma was generated at the position of the coil, and the plasma generator operated at a frequency of 13.56 MHz. The pristine Ga_2O_3 sample was placed 30 cm away from the plasma generator, minimizing ion bombardment damage to the film.

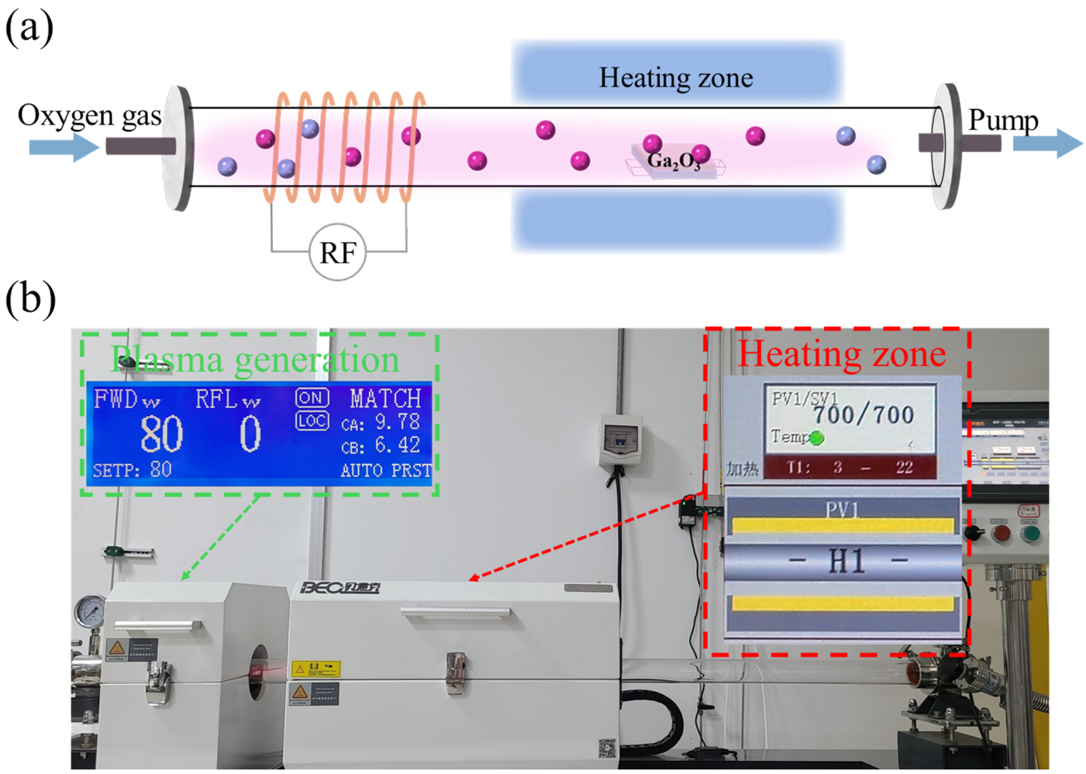

Figure S1. (a) Schematic diagram and (b) photograph of the remote plasma enhanced chemical vapor deposition system.

Supporting Note 2: Influence of different post-annealing temperatures on the photoelectric properties of Ga_2O_3-based SBPDs

In order to investigate the influence of different post-annealing temperatures on the photoelectric properties of Ga_2O_3-based SBPDs, we fabricated photodetectors based on Ga_2O_3 thin film annealed at different temperatures of 600, 700, 800 and 900 °C in an oxygen atmosphere, respectively. These four prepared photodetectors are designated as PD-600, PD-700, PD-800 and PD-900, respectively. The I-V characteristic curves measured in the dark and under 254 nm UV illumination with a voltage testing range from -20 to 20 V are shown in **Figure S2**a-d. We analyze the photocurrent and dark current at a bias of 20 V, summarized in **Figure S2**e. One can notice that, as the post-annealing temperature increases, the dark current gradually decreases, while the photocurrent sharply decreases after the post-annealing temperature exceeds 700 °C. Compared with PD-600, the photocurrent of PD-800 and PD-900 decreases by 96 and 4048 times, respectively. It is generally considered that oxygen vacancies in Ga_2O_3 thin film act as donor impurities, providing electrons and as such increasing carrier concentration.[1,2] As the post-annealing temperature increases, the concentration of oxygen vacancies in Ga_2O_3 decreases, and consequently, both photocurrent and dark current decrease. Although post-annealing leads to a simultaneous decrease in photocurrent and dark current, our goal is to obtain devices with high photocurrent and low dark current, and thus it is necessary to compare the performance of devices using the photo-to-dark current ratio (*PDCR*). We calculate and summarize the *PDCR* of these four devices at a bias of 20 V, as shown in **Figure S2**f. It can be seen that the *PDCR*s of PD-600, PD-700, PD-800, and PD-900 are 2.3×10^5, 6.2×10^5, 4.9×10^5, and 2.0×10^4, respectively. As the post-annealing temperature increases, the *PDCR* initially increases and then decreases, reaching its maximum value at 700 °C. Therefore, we conclude that the best device performance can be achieved at the post-annealing temperature of approximately 700 °C.

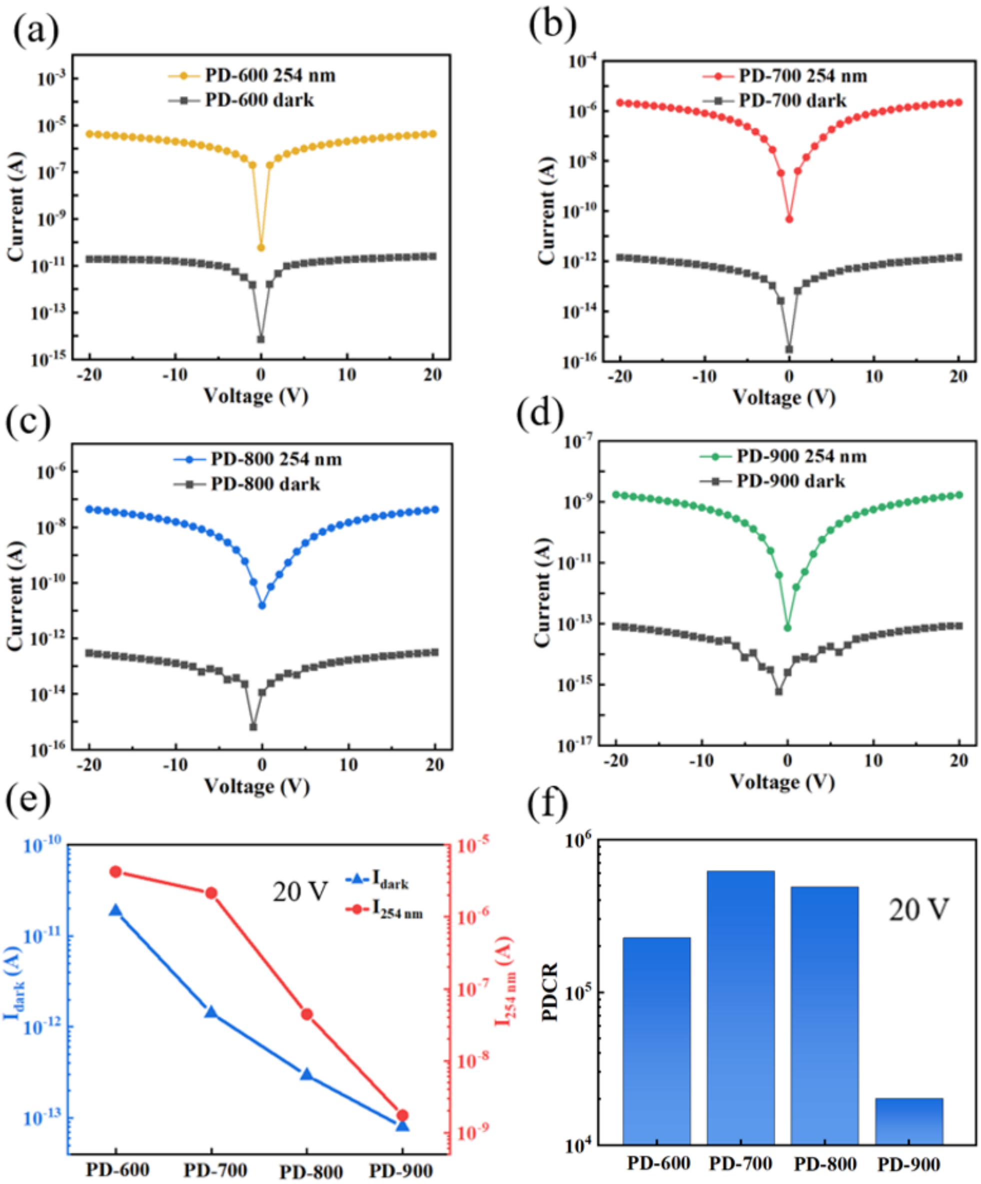

Figure S2. I-V characteristic curves of (a) PD-600, (b) PD-700, (c) PD-800, and (d) PD-900 in the dark and under 254 nm UV illumination; (e) Summary chart of the photocurrent and dark current at a bias of 20 V; and (f) Summary chart of the photo-to-dark current ratio (*PDCR*) at a bias of 20 V.

Supporting Note 3: Influence of plasma treatment at different processing

temperatures on the photoelectric properties of Ga_2O_3-based SBPDs

In order to investigate the influence of plasma treatment at different processing temperatures on the photoelectric properties of Ga_2O_3-based SBPDs, we fabricated four photodetectors based on Ga_2O_3 thin film treated with oxygen plasma at different processing temperatures of room temperature, 600, 700 and 800 ℃, respectively. These four prepared photodetectors are designated as PD-P-RT, PD-P-600, PD-P-700 and PD-P-800, respectively. The I-V characteristic curves measured in the dark and under 254 nm UV illumination with a voltage testing range from -20 to 20 V are shown in **Figure S3**a-d. We analyze the photocurrent and dark current at a bias of 20 V, summarized in **Figure S3**e. One can notice that, as the plasma treatment temperature increases, the photocurrent of the device decreases significantly at 800 ℃, while the decrease in the photocurrent is minor at 700 ℃ and earlier, remaining relatively stable. As the plasma treatment temperature increases, the dark current of the device gradually decreases, reaching 44 and 33 fA at 700 and 800 ℃, respectively. We summarize the photo-to-dark current ratio of these four devices at a bias of 20 V, as shown in **Figure S3**f. It can be seen that the *PDCR*s of PD-P-RT, PD-P-600, PD-P-700, and PD-P-800 are 5.1×10^5, 1.85×10^7, 4.7×10^7, and 1.32×10^6, respectively. The *PDCR* of the device is the highest at a plasma treatment temperature of 700 ℃, while a further increase in the plasma treatment temperature significantly decreases the photocurrent with minor decrease in the dark current, and thus reduces the *PDCR* at 800 ℃. Therefore, 700 ℃ is the optimal plasma treatment temperature.

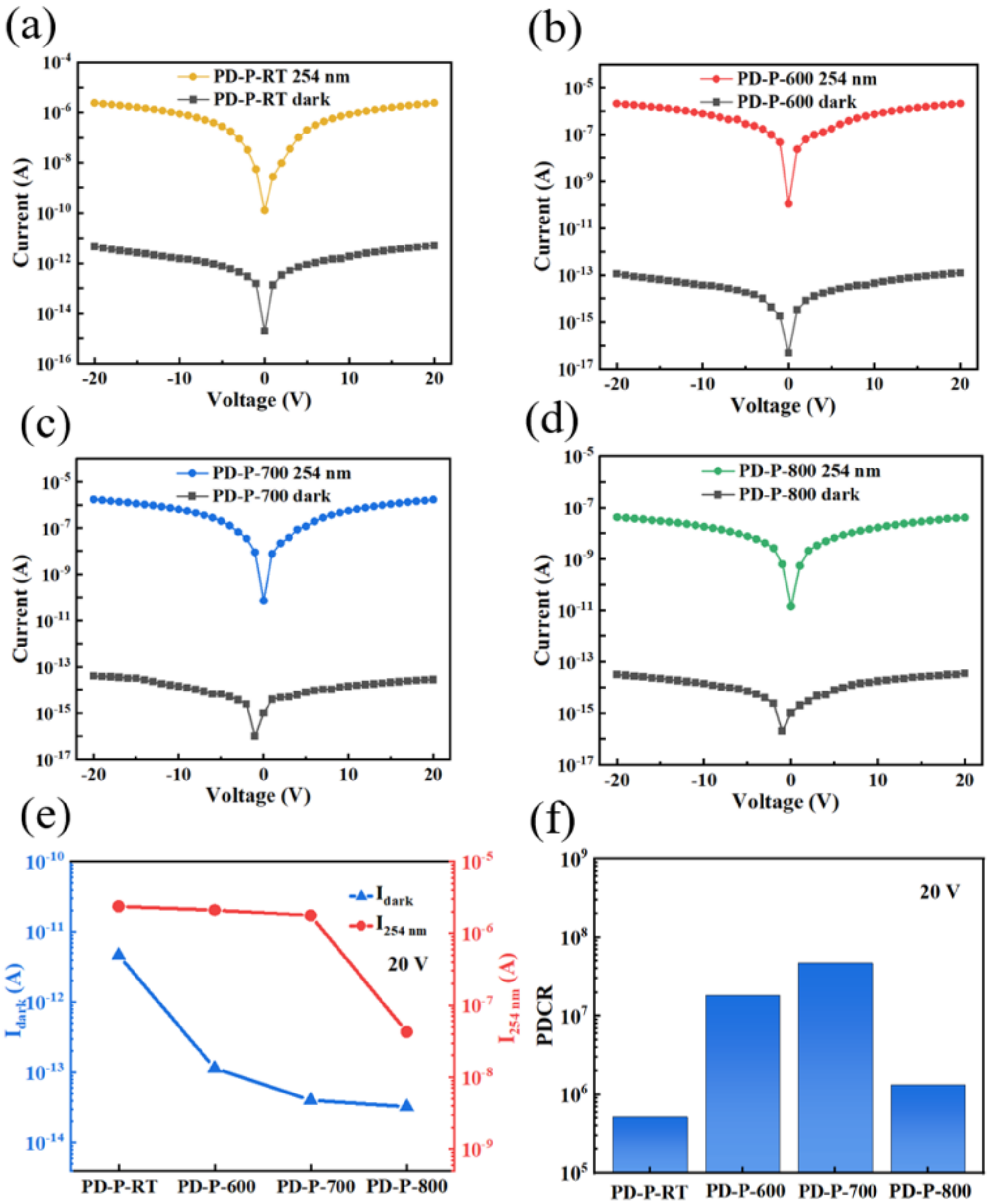

Figure S3. I-V characteristic curves of (a) PD-P-RT, (b) PD-P-600, (c) PD-P-700, and (d) PD-P-800 in the dark and under 254 nm UV illumination; (e) Summary chart of the photocurrent and dark current at a bias of 20 V; and (f) Summary chart of the photo-to-dark current ratio (*PDCR*) at a bias of 20 V.

Supporting Note 4: I-V characteristic curves of the prepared four SBPDs in the dark on a linear scale.

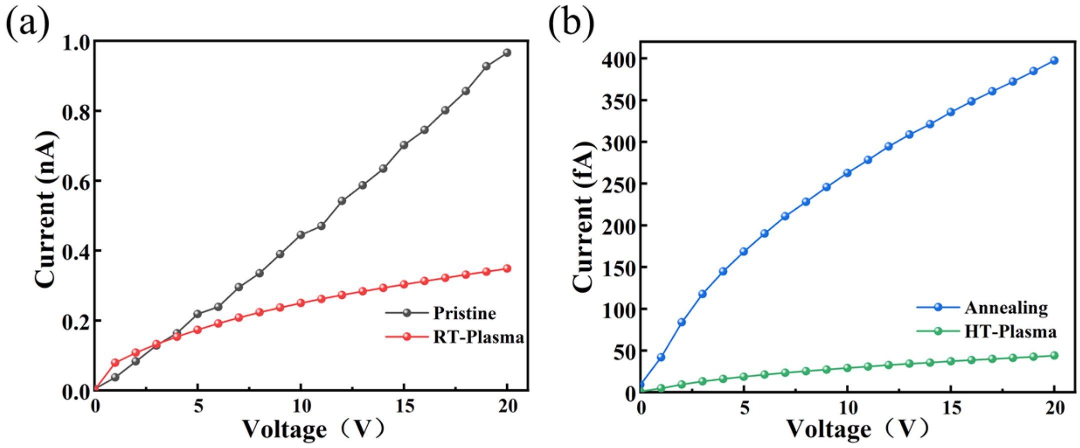

Figure S4. I-V curves of the MSM structured photodetectors based on (a) Pristine and RT-Plasma samples, (b) Annealing and HT-Plasma samples.

Figure S4a shows the I-V curves of the MSM structured photodetectors based on Pristine and RT-Plasma samples while **Figure S4**b shows the I-V curves of the MSM structured photodetectors based on Annealing and HT-Plasma samples in the dark on a linear scale. One can notice that: (i) the I-V curve for the photodetector based on the Pristine sample is almost linear, implying that the Ti/Cu metal electrodes form a quasi-Ohmic contact with the Ga_2O_3 thin film; (i) the I-V curves for the other photodetectors based on RT-Plasma, Annealing, and HT-Plasma samples exhibit significant nonlinearity, implying that the Ti/Cu metal electrodes form a Schottky contact with the Ga_2O_3 thin film.

Supporting Note 5: Performance of a single device in the photodetector array

To measure the performance of a single device in the photodetector array, we first selected one device for photoelectric performance testing, as depicted in **Figure S5**a. During the measurement, we placed one probe of the semiconductor parameter analyzer on the common electrode of all devices, and the other probe of the semiconductor parameter analyzer on the wire led out from the opposite electrode of the specific device, allowing the device to operate under an applied bias. The typical I-V characteristic curves of a single device in the array in the dark and under 254 nm UV illumination are shown in **Figure S5**b. The I-V characteristic curves of a single device's photocurrent and dark current both exhibit back-to-back Schottky rectifying characteristics, consistent with the feature of MSM structured photodetectors. The dark current of a single device at an applied bias of 20 V is below 0.1 pA, indicating a high impedance of a single device in the dark and low power consumption when the photodetector array is not operating. Upon exposure to 254 nm light, the single device's current significantly increases, with a photocurrent of 0.54 μA at a bias of 20 V, resulting in a *PDCR* of around 10^7. The extremely low dark current and high photo-to-dark current ratio enable the photodetector array to respond to weak light signals, suitable for applications requiring high-precision ultraviolet detection. The spectral response of a single device is shown in **Figure S5**c, with a peak response wavelength of 252 nm and low response in the wavelength range above 280 nm, demonstrating excellent spectral selectivity. **Figure S5**d illustrates the I-T characteristic curve of a single device with repeated switching of the device's current between the photocurrent and the dark current upon multiple opening and closing of the light source under a bias of 20 V and 254 nm UV illumination. The result of **Figure S5**d indicates the excellent repeatability of a single device in the photodetector array.[3]

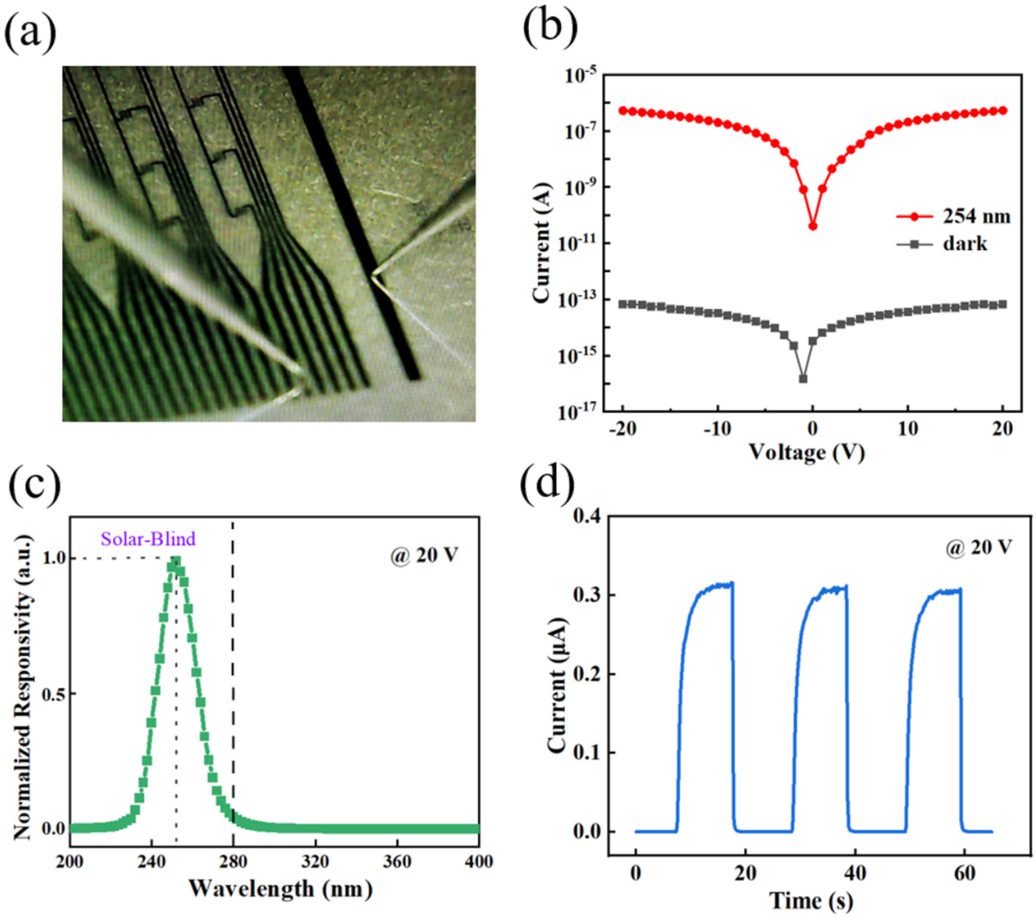

Figure S5. (a) Performance testing image of a single device in the photodetector array; (b) I-V characteristic curves of a single device in the photodetector array in the dark and under 254 nm UV illumination; (c) Normalized spectral response curve of a single device in the photodetector array at a bias voltage of 20 V; and (d) Multi-cycle I-T characteristic curves of a single device in the photodetector array at a bias voltage of 20 V.

References

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