Simultaneously improved sensitivity and response speed of $\beta$-Ga$_2$O$_3$ solar-blind photodetector via localized tuning of oxygen deficiency

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S1. Device fabrication

Firstly, the c-plane sapphire substrates were ultrasonically cleaned in acetone, ethanol, and deionized water successively. Then, a 100-nm $\beta$-Ga$_2$O$_3$ film was deposited on the sapphire substrates by plasma-assisted MBE using the optimized process conditions as follows: a base pressure of 1.0×10⁻⁹ Torr, Ga Knudsen cell temperature of 940 °C, substrate-heating temperature of 760 °C, input RF power of 300 W, and O$_2$ flow rate of 2 sccm. The MSM PDs were fabricated using the lift-off process. Each device comprised 20 pairs of interdigital electrodes whose length, width and spacing were 180 µm, 5 µm, and 5 µm, respectively; then, the effective illumination area was 3.51×10⁻⁴ cm². Moreover, each electrode was composed of 20-nm Ti and 80-nm Au, both grown by electron beam evaporation. The thin Ti layer was employed to improve the adhesion as well as the electrical contact of Au on underlying $\beta$-Ga$_2$O$_3$ film. Before the metal depositions, the area without the cover of photoresist was exposed to the Ar plasma (50 sccm) using a reactive-ion-etching system for 5 min with the different RF powers: 30 W and 60 W. Finally, all the PDs were annealed at 500 °C for 5 min in a N$_2$ ambient.

S2. Device measurements

For the material properties, the phase formation and crystallinity of $\beta$-Ga$_2$O$_3$ epitaxial films were monitored by Bede D1 XRD with monochromatic Cu Kα illumination ($\lambda = 1.5418$ Å); their optical transmission spectra were characterized by a PERSEE TU-1810 spectrophotometer; the surface morphologies of the $\beta$-Ga$_2$O$_3$ films with different Ar-plasma treatment
conditions were determined by a Seiko Instruments SPA-300 HV AFM; their elemental composition and chemical state were investigated by XPS using a Thermo Fisher K-Alpha system with an Al anode X-ray source (hν = 1456.5 eV). Regarding the device performance, the I-V and transient response were measured using an Agilent 4155B semiconductor parameter analyzer, with a low-pressure mercury lamp combined with a 254-nm filter as the UV-C light source (34.7 μW/cm²); the spectral response was measured using a Zolix DSR100-X150AUV automated spectro-radiometric measurement system. For the transient response measurement, an auto-controlled shutter was used to control the illumination with a trigger interval of 150 s. Zolix DSR100-X150AUV is composed of a UV enhanced xenon lamp (150 W), a monochromator, an optical chopper, a light-focusing lens assembly, a lock-in amplifier, and a probe station in dark chamber. The light from xenon lamp was chopped at 84 Hz and dispersed through the monochromator. Then, it was focused on the PD in dark chamber by the focusing lens assembly. The photocurrent was amplified and measured using a lock-in amplifier (Stanford SR830). The lamp power spectrum was calibrated using a standard UV-enhanced Si photodiode. Then, the output photocurrent was scaled with respect to the lamp power to yield the responsivity in A/W. The PDs were all under a 10-V bias during the transient and spectral response measurements.

S3. Out-of-plane XRD spectrum of β-Ga₂O₃ epitaxial film on c-plane sapphire substrate

![XRD spectrum of β-Ga₂O₃ epitaxial film on c-plane sapphire substrate](image)

Figure S1: XRD spectrum of β-Ga₂O₃ epitaxial film grown on c-plane sapphire substrate. All the peaks correspond to β-Ga₂O₃ (201) and the corresponding higher-order diffractions (located at 18.88°, 38.26°, and 58.96°), except those from the sapphire substrate (JCPDS CARD No. 43-1012), indicate a single-phase structure with (201) preferred orientation.

S4. Transmission spectrum of β-Ga₂O₃ epitaxial film on quartz glass substrate
Figure S2: Transmission spectrum of β-Ga$_2$O$_3$ epitaxial film on quartz glass substrate with the respective Tauc plot in the inset. The optical bandgap is determined by the Tauc method, that is, extrapolating the linear region of the incident photon energy (hv) versus (ahv)$^2$ to the horizontal axis. The absorption coefficient $\alpha$ is evaluated using the equation: $\alpha = 1/t \ln(1/T)$, where $t$ is the film thickness and $T$ is the film transmittance.

S5. AFM three-dimensional (3D) images of β-Ga$_2$O$_3$ epitaxial films

Figure S3: AFM 3D images of β-Ga$_2$O$_3$ epitaxial films with different Ar-plasma treatment conditions: (a) sample-ctrl, (b) sample-30, and (c) sample-60; (d) root-mean-square (RMS) versus Ar-plasma treatment condition.

6. I-V characteristics of the β-Ga$_2$O$_3$ MSM PDs in dark and under 254-nm illumination
Figure S4: Semi-logarithmic I-V plots of the $\beta$-Ga$_2$O$_3$ PDs: (a) in dark and (b) under 254-nm illumination; (c) responsivity and (d) specific detectivity versus Ar-plasma pretreatment condition.