

High-performance fully transparent Ga₂O₃ solar-blind UV photodetector with the embedded indium–tin–oxide electrodes

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ABSTRACT

Transparent ultraviolet (UV) photodetectors have attracted the increasing attention due to their giant potential in integrated transparent electronics applications. In this work, a fully transparent metal-semiconductor-metal (MSM) solar-blind UV photodetector with embedded indium–tin–oxide (ITO) electrodes based on Ga₂O₃ films was successfully designed and constructed for the first time. A novel method to prepare a MSM photodetector with embedded electrode structure is proposed by selective epitaxial growth of β -Ga₂O₃ thin films on *c*-plane sapphire substrate with ITO inter-digital electrodes prepared in advance. An ultra-low dark current of 1.6 pA, a superb UV-to-visible rejection ratio of 1.3×10^6 (R_{250}/R_{400}), a high specific detectivity of 7.4×10^{15} Jones and a large responsivity of 74.9 A/W can be observed in our device at 10 V, which are superior to those of other reported transparent UV photodetectors based on Ga₂O₃ films. The strong and uniform electrical field in β -Ga₂O₃ between two adjacent embedded ITO electrodes, and the high quality of β -Ga₂O₃ films should be responsible for the excellent solar-blind photodetection performance. Our findings pave a new way to realize a high-performance fully transparent Ga₂O₃ solar-blind UV photodetector, which has the giant potential for applications in future transparent electronics.

1. Introduction

Solar-blind ultraviolet (UV) photodetectors show numerous applications in various fields, including missile warning, environment monitoring, space research, flame detection, and so on [1–5]. Wide bandgap semiconductor materials, such as diamond, ZnMgO, AlGaIn and Ga₂O₃, have great potential for application in the UV photodetectors for their unique physical and chemical properties [6–11]. As the emerging wide bandgap semiconductor material, Ga₂O₃ is considered as an ideal material for the solar-blind photodetection, which benefits from its very suitable bandgap (4.7–5.3 eV), excellent chemical inertness, thermal stability and high radiation hardness [12–15]. To date, a large number of Ga₂O₃-based photodetectors with different structural types have been demonstrated, including metal–semiconductor–metal (MSM) structure, Schottky junction and heterojunction [16–19]. Among them,

owing to the fast response speed, simple fabrication process and easy integration with other devices, the inter-digitated MSM structure has become the most common architecture of Ga₂O₃ solar-blind UV photodetectors [14,20]. It is not uncommon to achieve large responsivity (>100 A/W) [21], high UV-to-Visible rejection ratio (>10⁵) [22], and high specific detectivity (>10¹⁴ Jones) [23] in Ga₂O₃ MSM photodetectors. Recently, transparent electronics, as a promising technology for next generation electronic and optoelectronic devices, have attracted increasing attention in various applications [24–27]. In particular, transparent UV photodetectors are crucial for integrated transparent electronics applications, such as smart windows, transparent display panels, transparent batteries, and so on [28–30]. However, for the most reported Ga₂O₃ MSM photodetectors, the opaque metals with high electrical conductivity have been employed as electrodes materials, which hinder applications of the devices in transparent electronics.

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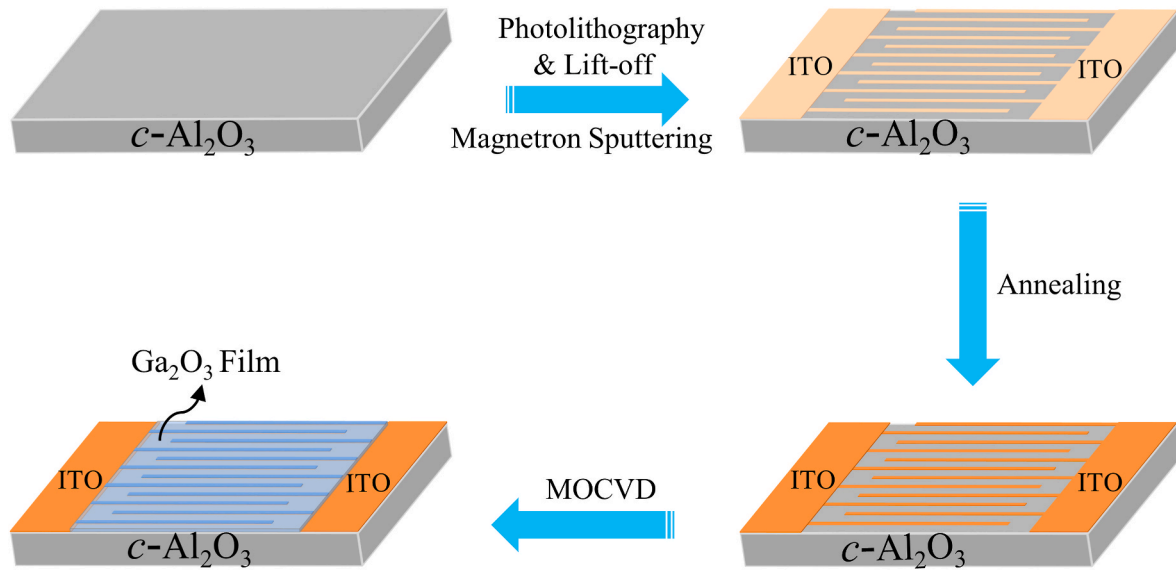


Fig. 1. Schematic diagram of fabrication processes of transparent Ga_2O_3 MSM photodetector.

Therefore, to meet the requirements of fully transparency, it is the best choice to replace the traditional metal electrodes with transparent electrodes in the UV photodetectors. So far, few reports on MSM-type Ga_2O_3 photodetectors with transparent electrodes are available [21, 31–33]. Kumar et al. reported a completely transparent and flexible amorphous Ga_2O_3 solar-blind UV device using amorphous indium-zinc-oxide ($a\text{-IZO}$) electrodes. The photodetector shows higher external quantum efficiency and responsivity compared to the device based on traditional Ag metal electrodes [31]. S. Oh et al. made the $\beta\text{-Ga}_2\text{O}_3$ film-based solar-blind photodetector using transparent graphene electrodes. In contrast to the device using Ni/Au metal electrodes, the transparent device exhibits higher performance, which is related to

the absence of shading and more carriers density with the graphene electrodes [32]. However, all the reported transparent Ga_2O_3 solar-blind UV photodetectors are fabricated by conventional planar electrode structures, whose inhomogeneous electric field within the active layer is detrimental to separation or collection of photogenerated electron-hole pairs [34,35].

In our work, Ga_2O_3 films were grown using metal–organic chemical vapor deposition (MOCVD) technology on $c\text{-Al}_2\text{O}_3$ substrates with interdigital indium tin oxide (ITO) electrodes prepared in advance, and a Ga_2O_3 MSM solar-blind UV photodetector with embedded electrodes was thus fabricated for the first time. The device shows a high transmittance (over 85% in 400–700 nm range), suggesting its fully

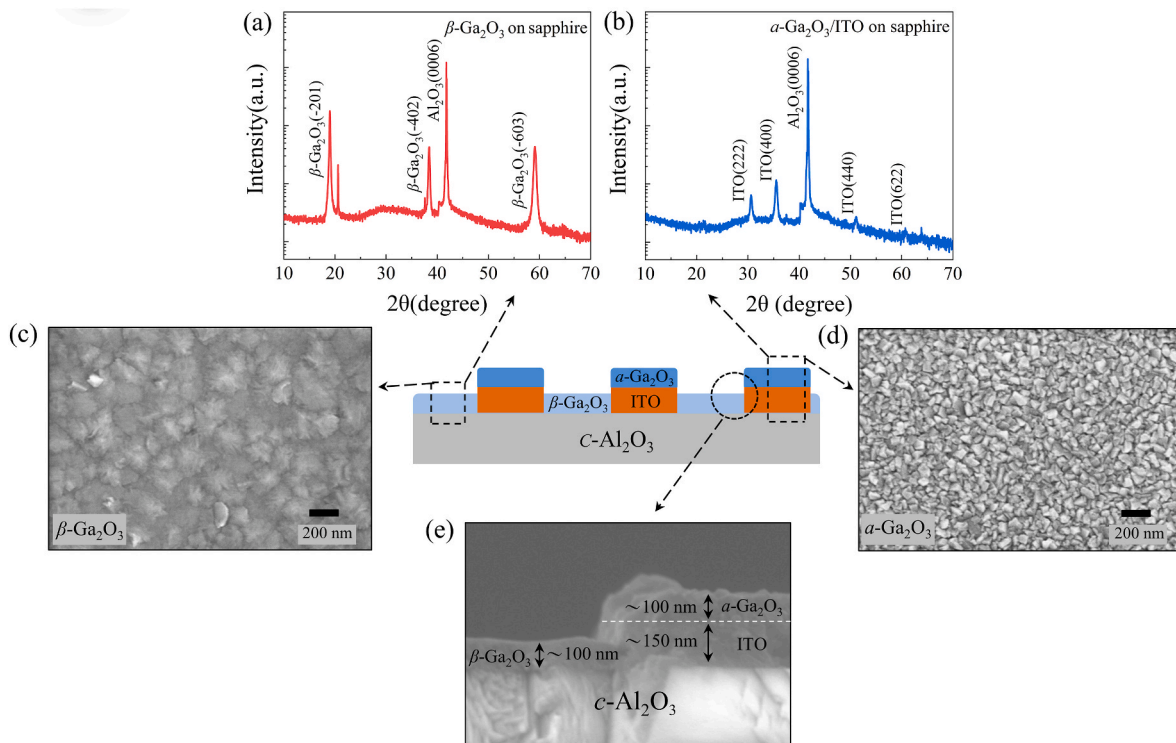


Fig. 2. XRD patterns of epitaxial Ga_2O_3 films (a) on $c\text{-Al}_2\text{O}_3$ substrate and (b) on ITO substrate. The top-view SEM images of Ga_2O_3 films (c) on $c\text{-Al}_2\text{O}_3$ substrate and (d) on ITO substrate. (e) The cross-sectional SEM image of device.

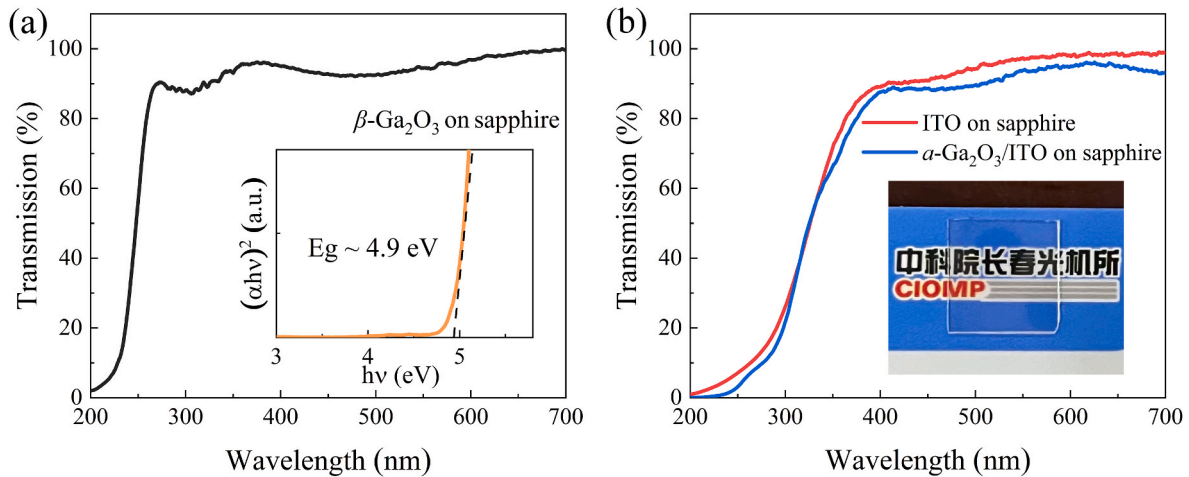


Fig. 3. Optical transmission spectra of (a) β -Ga₂O₃, (b) ITO and α -Ga₂O₃/ITO films. The inset in (a) is the Tauc's plots of film and the inset in (b) is original photograph of the device.

transparent nature. This transparent photodetector exhibits excellent solar-blind UV photodetection performance with an ultra-low dark current of 1.6 pA, a superb UV-to-visible rejection ratio of 1.3×10^6 (R_{250}/R_{400}), a large responsivity of 74.9 A/W and a high specific detectivity of 7.4×10^{15} Jones under 10 V bias. Moreover, the device also exhibits good stability and reproducibility with the quick response speed. The superb performance of our device with embedded electrodes can be attributed to more homogeneous electric field in Ga₂O₃ film, resulting in the enhancement of the separation and collection of photo-generated carriers. Our fully transparent Ga₂O₃-based MSM solar-blind UV photodetector has great application potential in integrated transparent electronics.

2. Experimental details

The schematic diagram of the fabrication processes of transparent Ga₂O₃ MSM photodetector was shown in Fig. 1. The ITO films was firstly deposited on *c*-plane sapphire substrate using magnetron sputtering with mixed gas flow rates of Ar (20 sccm) and O₂ (5 sccm) using the gas flow meters (pressure: ~ 1.3 Pa; power: 100 W). Subsequently, 25 pairs of ITO interdigital electrodes can be fabricated on sapphire by the photolithography & lift-off technique with ~ 500 μ m long, ~ 10 μ m wide and ~ 10 μ m spacing. Then ITO interdigital electrodes were annealed at 750 $^{\circ}$ C under the oxygen atmosphere for 40 min to obtain a low resistance ($\sim 1.54 \times 10^{-3}$ Ω cm of resistivity). After that, Ga₂O₃ films were grown on *c*-Al₂O₃ substrate with ITO interdigital electrodes by MOCVD, which were grown under the temperature of 700 $^{\circ}$ C with chamber pressure of 9.8×10^2 Pa. The gallium and oxygen precursors were Triethylgallium and high-purity oxygen with the flow rates of 4 and 500

sccm, respectively. Finally, transparent Ga₂O₃ MSM photodetector with embedded electrodes was demonstrated. The whole preparation process of our device does not require etching of Ga₂O₃, which not only simplifies the process, but also avoids the damage caused by etching.

The surface & cross-sectional morphologies of the films in our work were systematically characterized using scanning electron microscopy (SEM) (Hitachi S-4800) and crystalline structural properties of films were characterized using X-ray diffractometer (XRD) (Bruker D8GADDS). Transmission spectra were studied by the spectrophotometer (Shimadzu UV3101PC). For characterization of the current-voltage (*I*-*V*) and time-dependent current (*I*-*t*) were obtained using the semiconductor device analyzer (Agilent B1500A) at room temperature. The spectral response properties were measured using the UV-enhanced Xe lamp (200 W) with a monochromator.

3. Results and discussion

Fig. 2a exhibits XRD pattern of Ga₂O₃ film on *c*-Al₂O₃ substrate between ITO electrodes. Besides the diffraction peak from *c*-Al₂O₃, three peaks located at 18.88 $^{\circ}$, 38.26 $^{\circ}$, and 58.96 $^{\circ}$ can be attributed to the planes (-201), (-402) and (-603) of β -Ga₂O₃, respectively (JCPDS:43-1012). In contrast, for the Ga₂O₃ film on ITO, only the diffraction peaks of ITO and sapphire were observed, indicating that the Ga₂O₃ films grown on the ITO electrodes are amorphous as shown in Fig. 2b.

Additionally, top-view SEM images of β -Ga₂O₃ films on sapphire and amorphous Ga₂O₃ (α -Ga₂O₃) films on ITO are presented in Fig. 2c and d, respectively. Clearly, both two films have smooth surface and the grain size of β -Ga₂O₃ film is much larger than that of α -Ga₂O₃ film. At the same

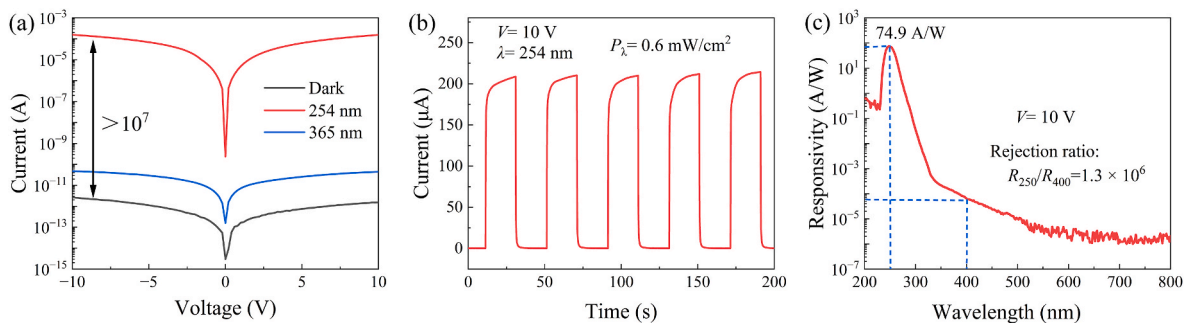


Fig. 4. (a) Semilogarithmic *I*-*V* characteristic curves of the device in dark and under UV light (254 and 365 nm). (b) *I*-*t* photoresponse curve with 254 nm illumination under 10 V bias. (c) Spectral response of the device under 10 V bias.

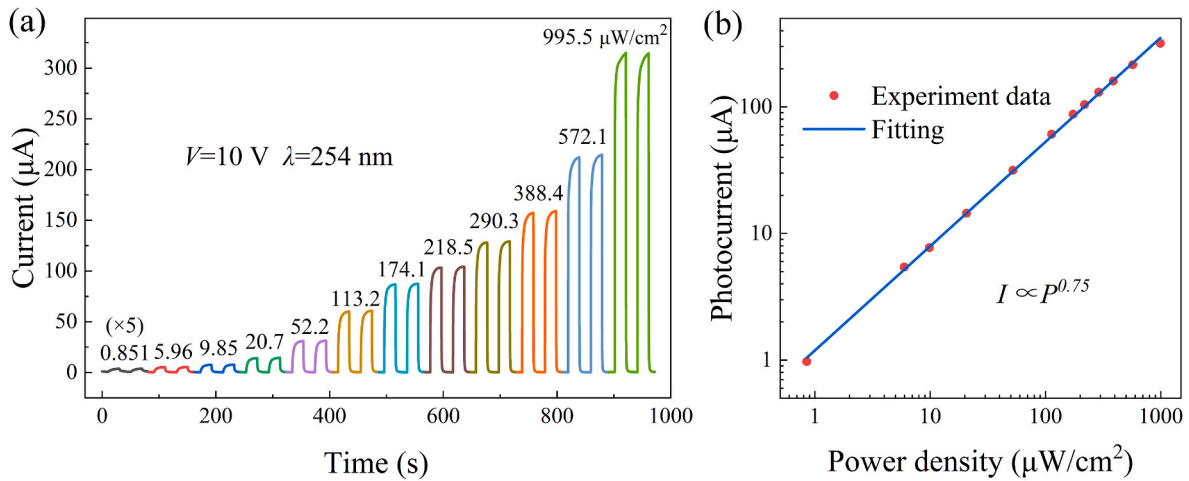


Fig. 5. (a) Time dependence of photocurrent characteristics of the device under various intensity 254 nm light illumination. (b) The photocurrent as the function of various illumination intensities in logarithmic form under 254 nm light biased at 10 V.

time, Fig. 2e displays the cross-sectional SEM image of the device, and we can see that both β -Ga₂O₃ and α -Ga₂O₃ films have the thickness of approximately 100 nm and ITO interdigital electrodes are approximately 150 nm thick. Therefore, the photosensitive area of our device should be the β -Ga₂O₃ film between two ITO electrodes, while α -Ga₂O₃ film on ITO has almost no contribution to photoresponse.

Fig. 3a presents optical transmission spectrum of β -Ga₂O₃ thin film on sapphire substrate, which shows a high transmittance (higher than 90%) in the spectral range of 400–700 nm. And a sharp absorption edge can be obtained at \sim 250 nm, corresponding to a \sim 4.9 eV bandgap (the inset of Fig. 3a). In addition, the transmittance of both ITO and α -Ga₂O₃/ITO films was more than 85% throughout the visible band as shown in Fig. 3b, which can meet the requirement of applications with high transparency. Furthermore, the actual photograph of the device (the inset of Fig. 3b) also confirms that it is almost fully transparent to the naked eyes and is very suitable for transparent electronics applications.

Fig. 4a shows the semi-logarithmic I - V characteristic curves of transparent Ga₂O₃ UV photodetector in dark and under UV (254 and 365 nm) light (Power density: \sim 0.6 mW/cm²) as shown in Fig. 4a. The transparent photodetector shows an ultra-low dark current (1.6 pA@10 V), suggesting the photodetector can be used for the detection of very weak light signals. The normalized photocurrent-to-dark current ratio (NPDR) is a objective metric for evaluating sensitivity of photodetectors, which can avoid the effect of input light intensities. And it can be

calculated using the following formula [36,37].

$$NPDR = \frac{I_{\text{photo}}/I_{\text{dark}}}{P_i S}$$

where the parameters include I_{photo} (photocurrent), I_{dark} (dark current), S (effective illumination area of devices) and P_i (power density of light). The calculated NPDR of our device is as high as $4.71 \times 10^{13} \text{ W}^{-1}$ under 254 nm light, indicating the excellent solar-blind UV photodetection performance. It is worth mentioning that our photodetector has an ultra-weak photoresponse to the 365 nm illumination, which indicates it has good suppression of the photoresponse from bands over solar-blind UV range. Fig. 4b shows the I - t curve of our device irradiated with 254 nm illumination at 10 V. The photodetector exhibits the fast response (\sim 1.63 s of 10–90% rise time; \sim 0.38 s of 90–10% decay time) to UV light with excellent repeatability and stability.

Fig. 4c shows spectral response curve of the photodetector on a semilogarithmic scale from 200 to 800 nm under 10 V. The maximum responsivity of the device appears at around 250 nm, reaching \sim 74.9 A/W. Meanwhile, the sharp -3 dB cutoff wavelength can be observed at \sim 260 nm, indicating a true solar blindness. Typically, UV-to-visible rejection ratio (R_{peak}/R_{400}) is the critical merits to analyze spectral selectivity of photodetector. It can be seen that the rejection ratio of our transparent Ga₂O₃ UV photodetector is 1.3×10^6 , suggesting the high spectral selectivity of our device.

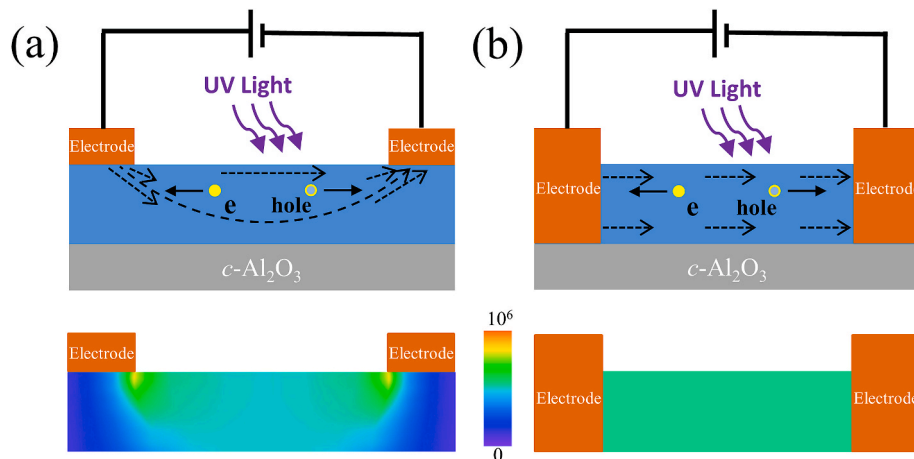


Fig. 6. Schematic diagrams and simulations of electric field distributions for Ga₂O₃ photodetectors with (a) planar structure and (b) embedded structure electrodes under 254 nm UV light illumination.

Table 1Performance comparison of our transparent device and recently reported typical MSM transparent photodetectors based on Ga₂O₃ thin film.

Materials	Electrode structure	Dark current (nA)	Responsivity (A/W)	Rejection ratio	Detectivity (Jones)	Ref.
α -Ga ₂ O ₃	Planar IZO	~1@200 V	3.2×10^{-4} @10 V	–	2.8×10^{10}	[42]
α -Ga ₂ O ₃	Planar AZO	2.84×10^{-3} @10 V	2.66@10 V	2.93×10^5 @10 V (R_{254}/R_{400})	4.84×10^{14}	[43]
α -Ga ₂ O ₃	Planar IZO	11@5 V	43.99@2 V	714.2@2V (R_{250}/R_{555})	–	[31]
α -Ga ₂ O ₃	Planar ITO	–	0.19@20 V	–	–	[44]
β -Ga ₂ O ₃	Planar ITO	3.8×10^{-3} @20 V	181.03@20 V	1.2×10^5 @20 V (R_{254}/R_{365})	1.2×10^{15}	[21]
β -Ga ₂ O ₃	Planar ITO	6×10^{-4} @10 V	–	6.83×10^3 (R_{250}/R_{400})	–	[45]
β -Ga ₂ O ₃	Embedded ITO	1.6×10^{-3} @10 V	74.9@10 V	1.3×10^6 @10 V (R_{250}/R_{400})	7.4×10^{15}	This work

Fig. 5a displays *I-t* photoresponse curves of our transparent Ga₂O₃ photodetector under 254 nm UV illumination of various intensities at 10 V. With the light density increasing ($0.851\text{--}995.5 \mu\text{W}/\text{cm}^2$), currents of device under light irradiation increase obviously with good stability and repeatability. In general, photocurrent (I_{photo}) versus power density (P) are fitted using following simple equation [38,39].

$$I_{\text{photo}} = AP^\theta$$

where A is a constant and θ is a exponent related to the trap states of the photodetector. As shown in Fig. 5b, θ value was estimated to be ~ 0.75 by fitting the data which is smaller than 1 (ideal trap-free device), suggesting the existence of the trap states in the Ga₂O₃ photodetector.

The specific detectivity (D^*) is also an critical merit to evaluate UV light detection ability of photodetectors, which is obtained from the following formula [40,41].

$$D^* = \frac{R}{\sqrt{2qI_{\text{dark}}/S}}$$

where R , q , I_{dark} and S correspond to the responsivity, the elemental charge, the dark current and the effective illumination area of devices, respectively. Obviously, the calculated D^* value of the photodetector is 7.4×10^{15} Jones, which benefits from its large responsivity and ultra-low dark current.

In order to investigate the reason for the excellent performance of our device, the simulations of electrical field intensity distributions for Ga₂O₃ photodetectors with planar and embedded electrodes were performed by the finite element method as shown in Fig. 6. Under 254 nm UV light illumination, the photogenerated carriers (electron-hole pairs) will be generated in photosensitive layer, which can be separated and moved towards to the electrodes by an applied electric field. It is obvious that for the device with planar electrode structure, the electric field density is greater in the region near the edges of electrodes, but smaller in the region between and under the electrodes. Conversely, when the electrodes are embedded in the β -Ga₂O₃ thin film, a more homogeneous and relatively stronger parallel electric field would be formed inside of the photosensitive layer, which is conducive to the separation and collection of photogenerated carriers. Therefore, the high performance of our transparent photodetector mainly comes from the embedded electrode structure. At the same time, the relatively high crystalline quality of β -Ga₂O₃ photosensitive layer and the device preparation process without etching are also very important factors.

Table 1 presents main performances of our device and some reported typical Ga₂O₃-based transparent photodetectors. Obviously, our device has a UV-to-visible rejection ratio (R_{peak}/R_{400}) of 1.3×10^6 and a specific detectivity of 7.4×10^{15} Jones, which are obviously larger than the recently reported Ga₂O₃-based transparent photodetectors. Additionally, the low dark current or peak responsivity of our transparent photodetector are very competitive or slightly better than the previously reported transparent Ga₂O₃ devices.

4. Conclusions

In summary, a high-performance fully transparent MSM structure solar-blind UV photodetector with embedded ITO electrodes was

constructed on Ga₂O₃ film by a novel method without any etching process. That is, the β -Ga₂O₃ film was directly and selectively grown on sapphire substrate with ITO interdigital electrodes prepared in advance. The photodetector exhibits an ultra-low dark current (1.6 pA), a high NPDR ($4.71 \times 10^{13} \text{ W}^{-1}$), a large responsivity (74.9 A/W), a superb rejection ratio (1.3×10^6) and a high specific detectivity (7.4×10^{15} Jones) at 10 V. In addition, the estimated rise and decay times are ~ 1.63 s and ~ 0.38 s, respectively. The comprehensive performance of our device is superior to previously reported transparent UV photodetectors based on Ga₂O₃ thin films, which can be attributed to its embedded electrode structure and high crystalline quality β -Ga₂O₃. Our work can provide a new method for the fabrication of fully transparent high-performance Ga₂O₃ solar-blind UV photodetectors.

Credit author statement

Chao Zhang: Investigation, Methodology, Formal analysis, Original draft preparation. Kewei Liu: Conceptualization, Validation, Formal analysis, Writing-Review & Editing, Supervision, Resources, Funding acquisition. Qiu Ai: Formal analysis, Resources, Supervision. Xuan Sun: Experimentation supporting. Xing Chen: Resources, Data curation. Jialin Yang: Resources, Data curation. Yongxue Zhu: Resources, Data curation. Zhen Cheng: Resources, Data curation. Binghui Li: Resources, Data curation. Lei Liu: Resources, Data curation. Dezhen Shen: Funding acquisition, Supervision, Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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