

# Self-powered solar-blind ZnGa<sub>2</sub>O<sub>4</sub> UV photodetector with ultra-fast response speed



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## ABSTRACT

A novel self-powered solar-blind ZnGa<sub>2</sub>O<sub>4</sub> ultraviolet (UV) photodetector has been constructed by fabricating asymmetric pairs of Au Schottky electrodes on the ZnGa<sub>2</sub>O<sub>4</sub> film. The current-voltage curve of the device exhibits an obvious rectifying characteristic with a rectification ratio of 11.3 at  $\pm 8$  V. At 0 V bias, the peak responsivity of our device at 246 nm is about 22.2 mA/W with an UV-vis rejection ratio of  $1.3 \times 10^4$ , indicating the excellent self-powered solar-blind UV photodetector performance. More interestingly, the rise time and decay time of the device are only 10 ns and 30 ns, respectively, which are much faster than that of any other previously reported self-powered solar-blind UV photodetectors to the best of our knowledge. The ultra-fast response speed of the device should be attributed to the asymmetric Schottky junctions and the high crystalline quality of our ZnGa<sub>2</sub>O<sub>4</sub> film with low trap density. Our work provides a new method to constructing high-performance self-powered solar-blind UV photodetectors.

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## 1. Introduction

Solar-blind ultraviolet (UV) photodetectors based on wide bandgap semiconductors (ZnMgO, AlGaN, diamond, Ga<sub>2</sub>O<sub>3</sub>, Zn<sub>2</sub>GeO<sub>4</sub>, ZnGa<sub>2</sub>O<sub>4</sub>, etc.) have drawn widespread attention due to their extensive applications in various fields, such as flame monitoring, missile warning, environmental monitoring, biological and chemical analysis, optical communication, space research and so on [1–8]. With the development of optoelectronic integration technology, there is an urgent need for high-performance UV detection devices with low energy consumption and high response speed. Therefore, the solar-blind UV photodetectors based on p-n junction and Schottky junction have been extensively explored due to the fact that they can work without external power source (known as “self-powered”) and have inherent advantages in sensitivity and response speed [9–13]. Unlike a p-n junction photodiode (a minority carrier device), a Schottky photodiode is known as a majority carrier device, which gives it a significant advantage in terms of response speed. In addition, the Schottky devices do not require p-type doping, which is extremely difficult for wide bandgap semiconductors [14–18]. Up to now, most of the reported self-powered

Schottky solar-blind UV detectors were based on vertical structures, and considerable progress has been achieved [19–22]. In particular, the responsivity of Au/ZnMgO/ZnO:Al vertical schottky photodiode at 266 nm can reach as high as 55 mA/W at 0 V with the rise and decay times of 0.02 ms and 0.3 ms, respectively [22]. In addition, the planar Schottky photodiode through different metal electrodes (one is an Ohmic contact and the other is a Schottky contact) was also demonstrated on diamond, which can be operated without any power supply. [23]. However, for these vertical and planar type Schottky photodetectors, both high-quality Ohmic and Schottky contacts are needed to the same semiconductor layer, which increases the difficulty in the preparation process. In addition, the response speed of the reported Schottky junction solar-blind UV photodetectors are still far from ideal, which is commonly considered to depend on the product of resistance and capacitance and the defect traps. Recently, a novel self-powered UV photodetectors based on an asymmetric metal-semiconductor-metal (MSM) structure (one metal interdigitated electrode with wide fingers and the other one with narrow fingers) has been first demonstrated on ZnO film by our group [24], and this idea was subsequently taken up by other researchers [25–28]. Compared with the conventional Schottky junction photodetectors, this novel asymmetric MSM photodetector has many special advantages, such as the simpler fabrication process, lower capacitance and weaker

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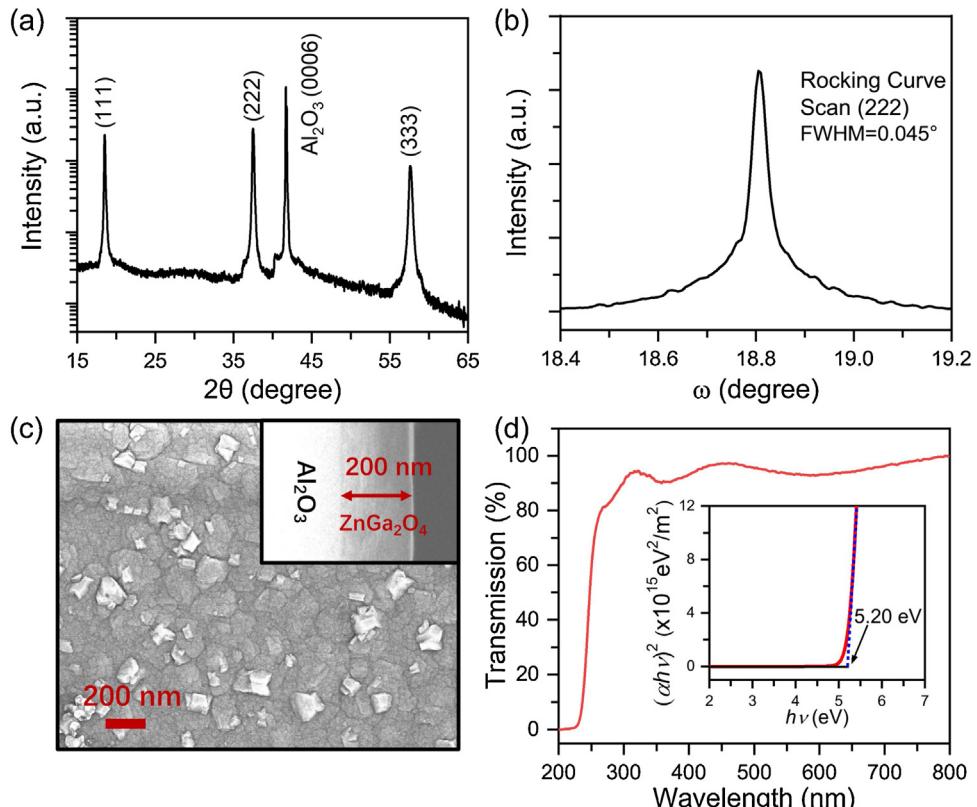
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light absorption by electrode, which could in turn result the higher responsivity and faster response speed.

In recent years, wide bandgap ternary oxides, such as  $\text{In}_2\text{Ge}_2\text{O}_7$  [29],  $\text{Zn}_2\text{GeO}_4$  [6,30] and  $\text{ZnGa}_2\text{O}_4$  [7,31], have received more and more attention in the fields of UV photodetection due to their tunable superior performance by altering the compositions. Among these ternary oxides,  $\text{ZnGa}_2\text{O}_4$  with a spinel structure has a wide bandgap of about 4.6–5.2 eV [32], directly corresponding to the solar-blind UV detection band with wavelength less than 280 nm. Unfortunately, almost all the reported  $\text{ZnGa}_2\text{O}_4$  solar-blind photodetectors were based on the traditional MSM structure, which cannot work without an external power supply [33–35]. In addition, although these devices demonstrated excellent performance in terms of responsivity, their response speed was not ideal. In this work, we constructed a self-powered solar-blind  $\text{ZnGa}_2\text{O}_4$  photodetector based on asymmetric pair of Au Schottky electrodes. The current-voltage ( $I-V$ ) curve of this novel self-powered solar-blind  $\text{ZnGa}_2\text{O}_4$  photodetector exhibits an obvious rectifying characteristic with a rectification ratio of 11.3 at  $\pm 8$  V. At 0 V bias, the peak responsivity of our device at 246 nm is around 22.2 mA/W with an UV-vis rejection ratio of  $1.3 \times 10^4$ , suggesting the excellent self-powered solar-blind UV photodetector performance. More interestingly, the rise time and decay time of the device are only 10 ns and 30 ns, respectively. To the best of our knowledge, our  $\text{ZnGa}_2\text{O}_4$  photodetector has the fastest response speed among the reported solar-blind photodetectors. Our findings in this work open up new ways to achieve high-performance self-powered solar blind UV detectors.

## 2. Experimental section

The  $\text{ZnGa}_2\text{O}_4$  film was epitaxially grown on *c*-face sapphire substrates by metal-organic chemical vapor deposition (MOCVD).



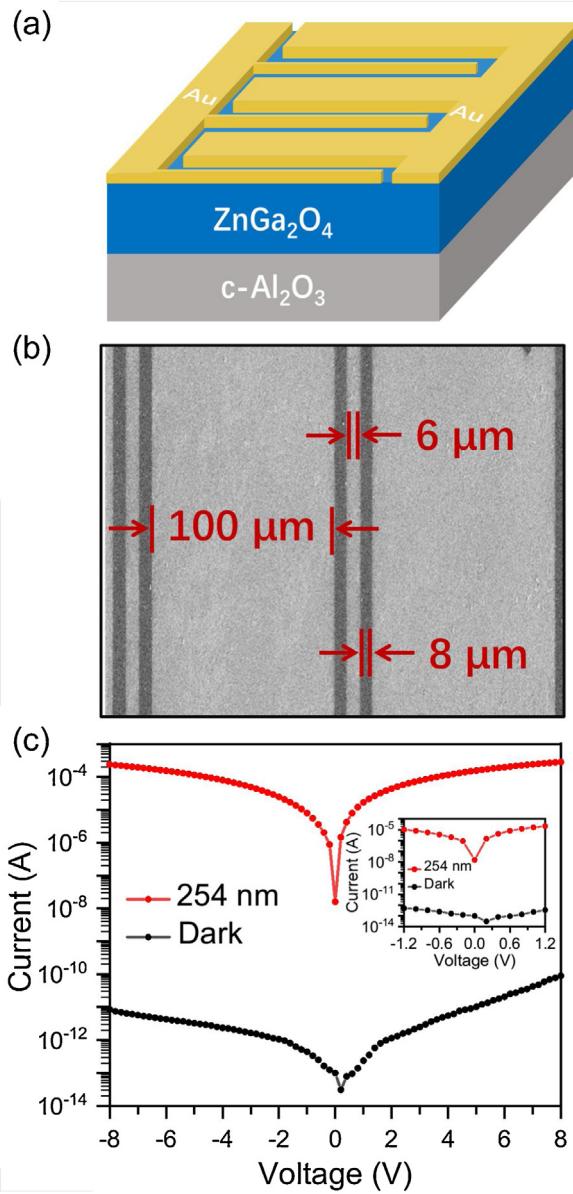
**Fig. 1.** (a) XRD pattern, (b) XRD rocking curve for (222) plane, (c) SEM images of surface and cross-sectional morphologies and (d) optical transmission spectrum of  $\text{ZnGa}_2\text{O}_4$  film. The inset of (d) shows a plot of  $(\alpha h\nu)^2$  as a function of photon energy ( $h\nu$ ).

Diethylzinc (DEZn), trimethylgallium (TMGa) and high-purity oxygen gas were used as the zinc, gallium and oxygen precursors, respectively. High-purity nitrogen gas was selected as the carrier gas. The film was grown at a substrate temperature of 850 °C with a chamber pressure of 23 Torr. The flow rates of DEZn, TMGa, and oxygen were kept at 20, 40 and 300 sccm, respectively. After the epitaxial growth of  $\text{ZnGa}_2\text{O}_4$ , the asymmetric Au interdigital electrodes were fabricated on the film by using the photolithography and lift-off technique to form MSM UV photodetector.

The optical, structural and morphology properties of  $\text{ZnGa}_2\text{O}_4$  film were characterized by adopting an UV-3101PC scanning spectrophotometer, a Bruker D8GADDS X-ray diffractometer (XRD) with Cu K $\alpha$  as the radiation source ( $\lambda = 0.154$  nm), and a scanning electron microscope (SEM) (HITACHI S-4800). The  $I-V$  curves and time-dependent photocurrent ( $I-t$ ) curves of the device were measured by a semiconductor device analyzer (Agilent B1500A). The transient response and spectral response of the device were measured by using an oscilloscope and a Nd:YAG laser (246 nm) and a 200 W UV-enhanced Xe lamp with a monochromator, respectively.

## 3. Results and discussion

Fig. 1(a) shows the XRD pattern of  $\text{ZnGa}_2\text{O}_4$  film. In Fig. 1(a), besides the diffraction peak from the *c*-plane sapphire substrate at 41.68°, three diffraction peaks at 18.50°, 37.48°, and 57.57° can be clearly obtained, corresponding to (111), (222) and (333) crystal facets of spinel  $\text{ZnGa}_2\text{O}_4$  (JCPDS No. 38-1240), respectively. Obviously, the (111) is the preferred growth orientation of the  $\text{ZnGa}_2\text{O}_4$  film on *c*- $\text{Al}_2\text{O}_3$ , which is consistent with the previous reports [36,37]. To further study the crystal quality of the  $\text{ZnGa}_2\text{O}_4$  film, the XRD rocking curve for the (222) plane of the sample was characterized as shown in Fig. 1(b). The full width at half maximum (FWHM) of the peak is only 0.045°, which is much narrower



**Fig. 2.** (a) Schematic illustration of the self-powered solar-blind ZnGa<sub>2</sub>O<sub>4</sub> MSM photodetector. (b) SEM image of asymmetric Au electrodes. (c) *I*-*V* characteristics of the device in dark and under 254 nm UV light illumination with an intensity of 1200  $\mu\text{W}/\text{cm}^2$ . The inset of (c) is the enlarged *I*-*V* characteristics near 0 V bias.

than that of the previously reported ZnGa<sub>2</sub>O<sub>4</sub> [38,39], indicating the higher crystalline quality of our ZnGa<sub>2</sub>O<sub>4</sub> film. Fig. 1(c) presents the SEM images of surface and cross-sectional morphologies of ZnGa<sub>2</sub>O<sub>4</sub> film. As shown in Fig. 1(c), the surface of ZnGa<sub>2</sub>O<sub>4</sub> film is relatively flat overall, and some granular structures can be found on the surface. And the thickness of ZnGa<sub>2</sub>O<sub>4</sub> film can be estimated to be about 200 nm by the cross-sectional SEM image (see the inset of Fig. 1(c)). Fig. 1(d) indicates the optical transmission spectrum of ZnGa<sub>2</sub>O<sub>4</sub> film, and it can be found that the average transmittance of the sample is over 90 % in the spectral range from 300 nm to 800 nm. In addition, a very sharp absorption edge can be clearly obtained at about 245 nm in the absorption spectrum, and the corresponding bandgap of the ZnGa<sub>2</sub>O<sub>4</sub> film is  $\sim 5.20$  eV, as shown in the set of Fig. 1(d).

Fig. 2(a) shows the schematic illustration of the self-powered solar-blind ZnGa<sub>2</sub>O<sub>4</sub> MSM photodetector. 12 pairs of asymmetric interdigital Au electrodes (50 nm thick) were defined by a con-

ventional photolithography and lift-off technique. The widths of wide Au electrode and narrow Au electrode are 100  $\mu\text{m}$  and 6  $\mu\text{m}$ , respectively, with 8  $\mu\text{m}$  gap and 500  $\mu\text{m}$  length, as shown in Fig. 2(b). To investigate the electrical and optoelectrical properties of the device, *I*-*V* characteristics measurements were carried out on the asymmetric ZnGa<sub>2</sub>O<sub>4</sub> MSM photodetector both in dark and under 254 nm UV light illumination with an intensity of 1200  $\mu\text{W}/\text{cm}^2$ . The forward bias direction of the device is defined as the application of forward voltage to the narrow Au electrode. As illustrated in Fig. 2(c), the contact between Au and ZnGa<sub>2</sub>O<sub>4</sub> is a typical Schottky contact, and an obvious rectifying characteristic with a rectification ratio of 11.3 at  $\pm 8$  V can be observed in dark. This phenomenon is in good agreement with our previously reported result [24]. In addition, the self-powered operations are observed from the enlarged *I*-*V* curves (inset of Fig. 2(c)) with an obvious photocurrent at zero bias under 254 nm UV light illumination with an intensity of 1200  $\mu\text{W}/\text{cm}^2$ .

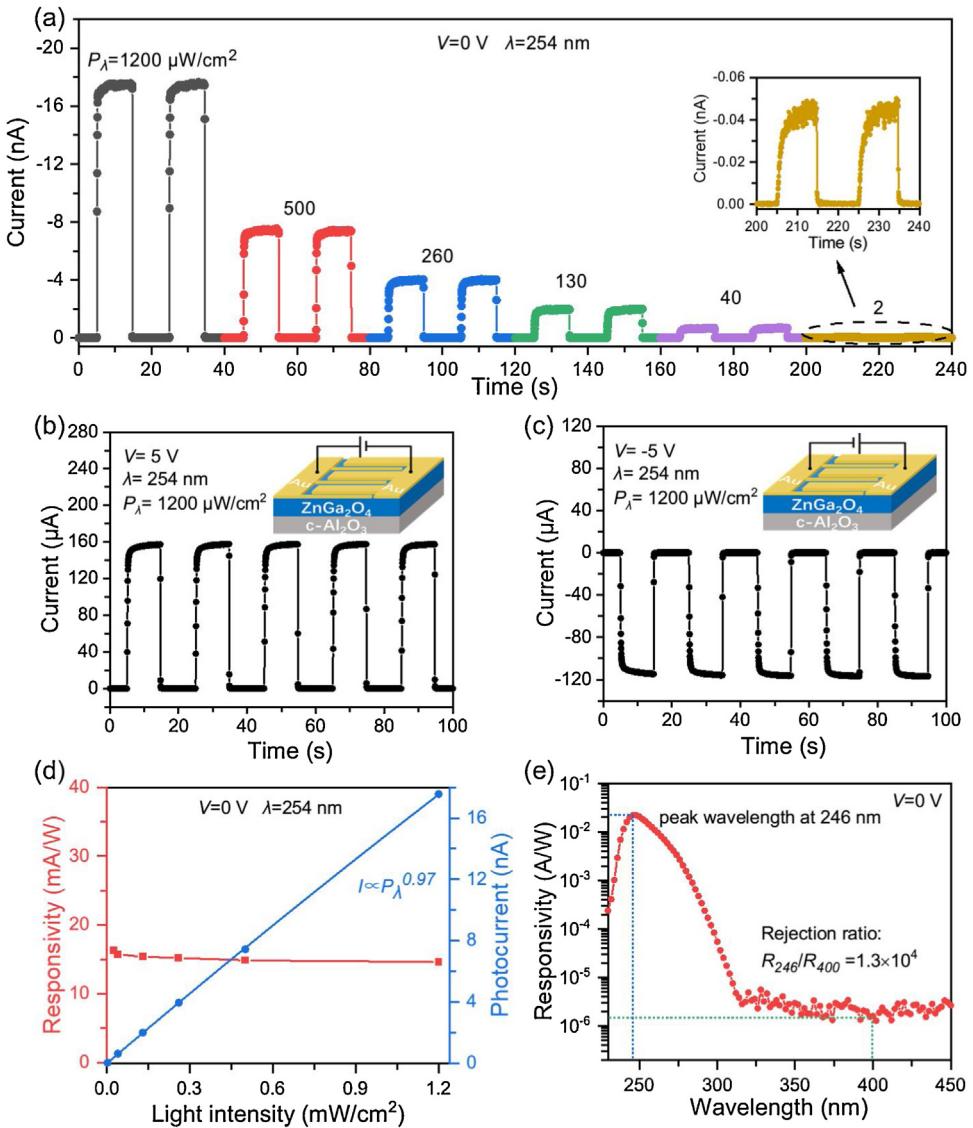
The photoresponse of the self-powered solar-blind ZnGa<sub>2</sub>O<sub>4</sub> MSM photodetector to 254 nm light illuminations has been systematically investigated by varying the power densities from 2  $\mu\text{W}/\text{cm}^2$  to 1200  $\mu\text{W}/\text{cm}^2$  at 0 V bias, as shown in Fig. 3(a). Obviously, the photodetector has a good ON/OFF switching performance with high reproducibility and stability under different light power densities. Even under low-intensity (2  $\mu\text{W}/\text{cm}^2$ ) light, our device still has good stability and obvious photocurrent ( $\sim 0.04$  nA), showing the excellent detection performance in solar-blind UV region [31,40]. Meanwhile, the rise and decay times of the device were very short, which even exceeded the measuring limit of a test apparatus (40 ms). Moreover, the similar good ON/OFF switching performance with high reproducibility and stability was also observed at 5 V and -5 V bias under 254 nm UV light illumination with an intensity of 1200  $\mu\text{W}/\text{cm}^2$  as shown in Fig. 3(b) and (c), respectively. The difference in photocurrent values of the device at  $\pm 5$  V is caused by the asymmetric pairs of Au Schottky electrodes.

Fig. 3(d) shows the photocurrent and the responsivity of the device under 254 nm illuminations with different light intensities at 0 V bias. As the incident light intensity increases, the photocurrent increases linearly. According to the previous reports, the relationship between the photocurrent and the light intensity can be described by a power law,  $I_{\text{ph}} \propto P^\alpha$ , where  $P$  is the light intensity and  $\alpha$  reflects the photocurrent efficiency [10]. By fitting the data, the  $\alpha$  can be estimated to be 0.97, which is very close to 1, suggesting the low trap density in the ZnGa<sub>2</sub>O<sub>4</sub> film [12,41]. The responsivity of the device is an important parameter of photodetector performance, which was calculated by the following formula [42]:

$$R = (I_{\text{photo}} - I_{\text{dark}})/PS$$

where  $I_{\text{photo}}$  is the photocurrent,  $I_{\text{dark}}$  is the dark current,  $P$  is the light intensity, and  $S$  is the effective area (the gap between the Au metal fingers) under irradiation. As shown in the Fig. 3(d), the responsivity decreases slightly with increasing the light intensity, which may be related to the relatively higher recombination rate of photogenerated carriers generated under stronger light intensity conditions [43].

Fig. 3(e) shows the spectral response of the self-powered solar-blind ZnGa<sub>2</sub>O<sub>4</sub> MSM photodetector at 0 V bias with  $y$  axis in logarithmic scale. Obviously, the peak responsivity occurs at 246 nm with -3 dB cut-off wavelength of 254 nm, which is in good agreement with the UV-vis transmission spectrum of the ZnGa<sub>2</sub>O<sub>4</sub> film. Only 8 nm difference between the response peak and -3 dB cut-off edge suggests that the device has excellent intrinsic solar-blind photodetection characteristics. The peak responsivity at 246 nm can reach to 22.2 mA/W. Moreover, the UV-vis rejection ratio, defined as the ratio between the peak responsivity and responsiv-



**Fig. 3.** (a) Time-dependent photocurrent characteristics of the self-powered solar-blind ZnGa<sub>2</sub>O<sub>4</sub> MSM photodetector under 254 nm illuminations with different light intensities from 2 to 1200  $\mu\text{W}/\text{cm}^2$  at 0 V bias. The inset of (a) is the enlarged  $I$ - $t$  curves under the corresponding illumination conditions. (b) Time-dependent photocurrent characteristics of the device at 5 V bias under 254 nm UV light illumination with an intensity of 1200  $\mu\text{W}/\text{cm}^2$ . The inset of (b) is the schematic diagram of the power supply connections of the device at 5 V bias. (c) Time-dependent photocurrent characteristics of the device at -5 V bias under 254 nm UV light illumination with an intensity of 1200  $\mu\text{W}/\text{cm}^2$ . The inset of (c) is the schematic diagram of the power supply connections of the device at -5 V bias. (d) The photocurrent and the responsivity of the device under 254 nm illuminations with various light intensities at 0 V bias. (e) The spectral response of the self-powered solar-blind ZnGa<sub>2</sub>O<sub>4</sub> MSM photodetector at 0 V bias with y axis in logarithmic scale.

ity at 400 nm, is up to  $1.3 \times 10^4$ . The Noise Equivalent Power (NEP) is the common metric that quantifies a photodetector's sensitivity (the detection limit) [44–49]. NEP defines the incident optical power required to obtain a signal-to-noise ratio (SNR) of 1, and it can be estimated by the following formula [50,51]:

$$\text{NEP} = \sqrt{4kT\Delta f/R_0}/R$$

where  $k$  is the Boltzmann's constant,  $T$  is the temperature,  $\Delta f$  (commonly 1 Hz) is a specified bandwidth, and  $R_0$  is the differential resistance. Under 0 V bias and at room temperature, the NEP of the self-powered solar-blind ZnGa<sub>2</sub>O<sub>4</sub> photodetector is around  $3.1 \times 10^{-15}$  W/ $\sqrt{\text{Hz}}$ .

Response time is another vital performance parameter for photodetectors. To precisely measure this parameter, the rise and decay times of the photodetector were measured using an oscilloscope and a 246 nm wavelength pulsed laser source. Fig. 4(a) shows the schematic diagram of the experimental setup used to measure the

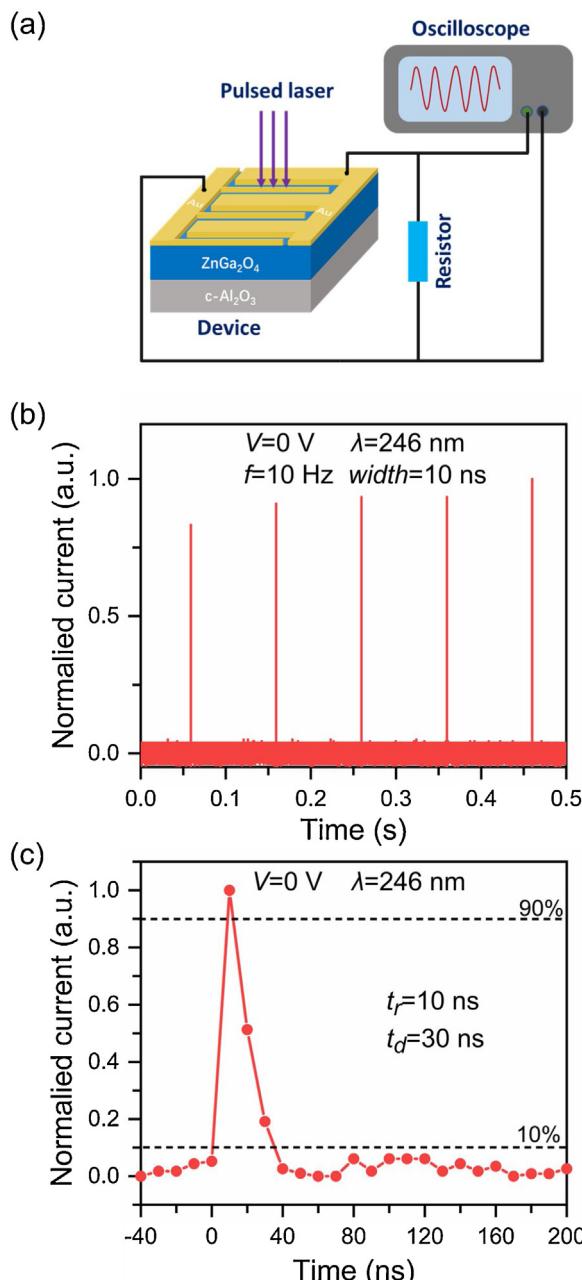
response time of ZnGa<sub>2</sub>O<sub>4</sub> photodetector. The laser pulse width and the frequency are 10 ns and 10 Hz, respectively. Fig. 4(b) shows the transient photoresponse of the self-powered solar-blind ZnGa<sub>2</sub>O<sub>4</sub> MSM photodetector at 0 V bias. As shown in Fig. 4(b), the photoresponse of our self-powered device is very fast, highly stable and reproducible. The rise and decay times, defined as the time for the current from 10 % to 90 % and from 90 % to 10 % of the peak value, can be estimated to be about 10 ns and 30 ns, respectively (see Fig. 4(c)). In this work, the rise time should be limited by the pulsed width of the laser. And the asymmetric Schottky junctions and the high crystalline quality of our ZnGa<sub>2</sub>O<sub>4</sub> film with low trap density can be responsible for the short decay time.

Table 1 summarizes the performance parameters of the reported typical self-powered solar-blind photodetectors. Obviously, the response speed of our device ( $t_r = 10$  ns,  $t_d = 30$  ns) is much faster than that of any other previously reported self-powered solar-blind UV photodetectors. Additionally, the UV-vis rejection ratio and

**Table 1**

The performance parameters of the reported typical self-powered solar-blind photodetectors.

Devices	Structure	Rise time/Decay time	Peak responsivity at 0 V ( $R_p$ )	Cut-off wavelength	Rejection ratio ( $R_p/R_{400}$ )	Ref.
$\beta\text{-Ga}_2\text{O}_3/\text{NSTO}$	Heterojunction	0.21 s/0.07 s	2.6 mA/W @ 254 nm	–	–	[52]
ZnO/ $\text{Ga}_2\text{O}_3$	Heterojunction	100 $\mu\text{s}/900 \mu\text{s}$	9.7 mA/W @ 251 nm	266 nm	$6.9 \times 10^2$	[15]
Polyaniline/MgZnO	Heterojunction	<0.3 s/<0.3 s	0.16 mA/W @ 250 nm	271 nm	$\sim 10^4$	[16]
$\text{MoS}_2/\beta\text{-Ga}_2\text{O}_3$	Heterojunction	–/–	2.05 mA/W @ 245 nm	260 nm	$1.6 \times 10^3$	[53]
Diamond/ $\beta\text{-Ga}_2\text{O}_3$	Heterojunction	–/–	0.2 mA/W @ 244 nm	270 nm	140	[17]
$\beta\text{-Ga}_2\text{O}_3/\text{Polyaniline}$	Heterojunction	0.34 ms/8.14 ms	21 mA/W @ 246 nm	272 nm	$3 \times 10^2$	[18]
Au/ZnMgO/ZnO:Al	Schottky junction	0.02 ms/0.3 ms	55 mA/W @ 265 nm	283 nm	–	[22]
Au/ $\beta\text{-Ga}_2\text{O}_3$	Schottky junction	1 $\mu\text{s}/64 \mu\text{s}$	0.01 mA/W @ 258 nm	270 nm	38	[19]
Au#1-ZnGa <sub>2</sub> O <sub>4</sub> -Au#2	asymmetric MSM	10 ns/30 ns	22.2 mA/W @ 246 nm	254 nm	$1.3 \times 10^4$	This work



**Fig. 4.** (a) Schematic diagram of our experimental setup for measuring the response time of the ZnGa<sub>2</sub>O<sub>4</sub> photodetector. (b) Transient photoresponse of the self-powered solar-blind ZnGa<sub>2</sub>O<sub>4</sub> MSM photodetector at 0 V bias under illumination of a Nd:YAG laser with a wavelength of 246 nm. (c) A single normalized cycle response of the device.

the peak responsivity of our device are higher than that of most reported self-powered solar-blind UV photodetectors at 0 V. It is worth noting that the asymmetric interdigital ZnGa<sub>2</sub>O<sub>4</sub> photodetector has an excellent repeatability. Different devices fabricated by the same process showed the similar self-powered solar-blind photodetection performance.

#### 4. Conclusions

In summary, a novel self-powered solar-blind UV photodetector was first constructed on ZnGa<sub>2</sub>O<sub>4</sub> film. A typical photovoltaic characteristic can be observed in the device at 0 V bias through a simple fabrication of the asymmetric pairs of Au planar electrodes. The  $I-V$  curve of this ZnGa<sub>2</sub>O<sub>4</sub> photodetector exhibits an obvious rectifying characteristic with a rectification ratio of 11.3 at  $\pm 8$  V. At 0 V bias, the response peak with a responsivity of 22.2 mA/W is situated at 246 nm with a -3 dB cut-off edge at 254 nm. And the UV-vis rejection ratio of our device is up to  $1.3 \times 10^4$ , indicating the device is a self-powered solar-blind UV detection with outstanding performance. Moreover, the rise time and decay time of the device are about only 10 ns and 30 ns, respectively, which are much faster than that of any other previously reported self-powered solar-blind UV photodetectors. Our work in this paper provides a new method to constructing high-performance self-powered solar-blind photodetectors.

#### CRediT authorship contribution statement

**Dongyang Han:** Investigation, Writing - original draft. **Kewei Liu:** Validation, Formal analysis, Writing - review & editing, Supervision, Resources. **Qichao Hou:** Investigation. **Xing Chen:** Resources, Visualization. **Jialin Yang:** Supervision. **Binghui Li:** Resources, Supervision. **Zhenzhong Zhang:** Resources, Supervision. **Lei Liu:** Resources, Supervision. **Dezhen Shen:** Writing - review & editing, 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.

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## Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.sna.2020.112354>.

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