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Self-powered solar-blind ultraviolet photodetector based on Au/ZnMgO/ZnO:Al with comb-shaped Schottky electrode

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ABSTRACT

We have demonstrated a self-powered solar-blind ultraviolet (UV) photodetector based on the Au/ZnMgO/ZnO:Al vertical structure. The comb-shaped Au was selected as the Schottky electrode due to its high transmittance (more than 60%) in the wavelength region from 200 nm to 300 nm. At 0 V bias, the detector showed a responsivity of 55 mA/W under 265 nm light illumination with the intensity of 80 nW/cm^2, which is larger than that of any other previously reported ZnO-based self-powered UV photodetectors. And a fast response speed can be also clearly observed with 10%–90% rise and 90%-10% decay times of 20 μs and 300 μs, respectively. The excellent self-powered solar-blind UV detection of our Au/ZnMgO/ZnO:Al device should be associated with the high external quantum efficiency, especially for the weak signal detection. Our findings offered new chances in high-performance self-powered photodetectors fabrication.

1. Introduction

Recently, solar-blind photodetectors operating in the spectrum region of 220 nm – 280 nm have been attracting more and more attention due to their huge potential for applications in fire monitoring, missile early warning, confidential communication, space exploration, etc [1–5]. In the last few decades, several kinds of wide bandgap semiconductors, such as GaAlN, ZnMgO, Ga_2O_3, and diamond, have been widely investigated and successfully used to fabricate intrinsic solar-blind ultraviolet (UV) photodetectors [6–10]. Among these materials, ZnMgO alloy is considered as one of the most promising candidates for solar-blind UV detection due to its excellent material properties, including tunable direct band gap, low defect density, environmental friendliness and strong radiation hardness, etc [7,11–13]. Recently, energy conservation becomes one of the most important issues around the whole world. Therefore, the self-powered devices have attracted more and more attention due to that they can be operated without any external power supply. As is well known, typical self-powered photodetectors are usually based on the PN and Schottky junctions due to the built-in electric field at the junctions [2,4,5,14–17]. Owing to the lack of reliable p-type materials, no information about ZnMgO homojunction solar-blind photodetectors can be found [7,18,19]. Although combining n-type ZnMgO alloy with other p-type layers (Si, NiO, SiC, etc.) could realize the self-powered devices, they always suffer from the unexpected visible/IR response from p-type materials and the lattice-mismatched heteroepitaxy [20–23]. Therefore, the Schottky junction seems like an ideal structure for the fabrication of ZnMgO solar-blind photodetectors. Endo, et al. reported a self-powered Pt/ZnMgO Schottky photodiode on ZnO single crystal substrate with the peak response of 15 mA/W at 220 nm under 0 V bias [27]. However, the high price of ZnO single crystal limits the further development of the devices based on this substrate. To solve this problem, ZnO buffer layer is usually introduced between a conventional substrate and ZnMgO film instead of a ZnO substrate [24–26]. Recently, our group demonstrated an Ag/ZnMgO Schottky solar-blind UV detector on sapphire substrate by introducing a thin ZnO buffer layer, and its peak responsivity is 16 mA/W at 278 nm under 0 V bias [4]. And the rise time and decay time were around 24 μs and 300 μs, respectively. For practical applications, high photosensitivity and quick response speed of UV photodetectors should be the pre-requisites. As is well known, the light usually passes through the Schottky electrode to reach the photosensitive area, and thus the light absorption of the elec-

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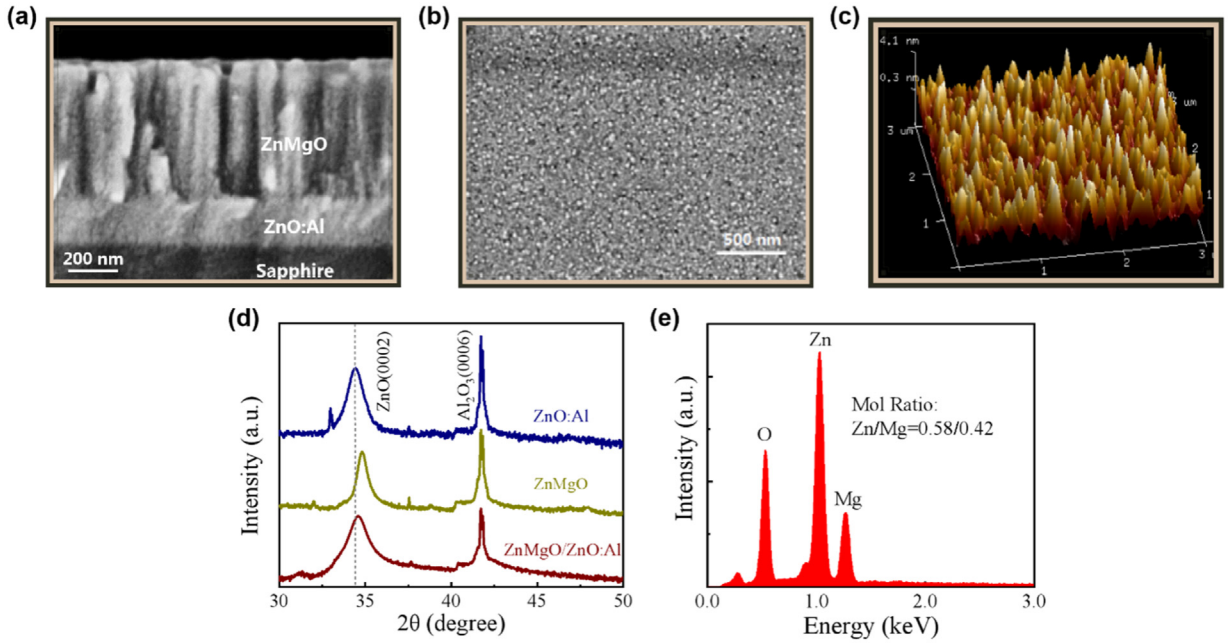

Fig. 1. (a) Cross-sectional SEM image, (b) Surface SEM image and (c) AFM image of the ZnMgO/ZnO:Al/sapphire, (d) XRD patterns of ZnO:Al/sapphire, ZnMgO/sapphire and ZnMgO/ZnO:Al/sapphire, (e) EDS spectrum of the ZnMgO film on sapphire substrate.

trode would strongly affect the performance of the Schottky UV photodetectors, especially for the weak signal detection. According to the previous reports, all the ZnMgO Schottky solar-blind UV photodetectors have selected metal film as Schottky contact electrode [4,6,7,14,27,28]. which would seriously reduce the number of photons reaching the photosensitive area. To resolve this problem, in this work, a comb-shaped Au Schottky electrode was fabricated on ZnMgO film to form an Au/ZnMgO solar-blind UV photodetector. At 0 V bias, the device showed a noticeable UV response with a response peak at 265 nm and a -3 dB cut-off edge at 283 nm. And 10%–90% rise and 90%-10% decay times are around 20 μs and 300 μs, respectively. In addition, with the decrease of the incident light power (λ = 265 nm), the responsivity increased obviously and could reach 55 mA/W with the 80 nW/cm^2 intensity, indicating excellent detection of weak light signals.

2. Material and methods

The ZnO:Al films were grown on *c*-Al_2O_3 substrates by radio frequency (RF) magnetron sputtering at 300 °C. Al (99.99%) and ZnO (99.99%) targets were sputtered in a reactive ambient of oxygen (20 sccm) and argon (20 sccm) mixture with a pressure of 3 Pa. The sputtering powers are at 100 W for both Al and ZnO targets. After the growth, the samples were annealed under 500 °C in the air by tube furnace to form ZnO:Al with low resistance. Then, the ZnMgO layer was grown on ZnO:Al layer by plasma-assisted molecular beam epitaxy (p-MBE). And 6N-purity zinc and 5N-purity magnesium held in thermal Knudsen cells and 5N-purity O_2 activated in a radio frequency plasma source were selected as precursors. During ZnMgO film growing, the chamber pressure was maintained at 10^{-3} Pa and the substrate temperature was kept at 200 °C. The radio frequency power was fixed at 300 W with O_2 flow rate of 1.2 sccm. In addition, we kept the temperature of Zn and Mg source at 230 °C and 352 °C, respectively. The surface and thickness of the film are evaluated by scanning electron microscopy (SEM). The structural property of the film is characterized by X-ray diffractometer (XRD) (Cu-Kα radiation, λ =0.154 nm). The surface roughness of ZnMgO film was estimated by atomic force microscopy (AFM; Bruker, MultiMode-8). The composition of the ZnMgO film was measured by energy dispersive X-ray spectrometer (EDS, GENESIS 2000 XMS60S) The electrical characteristics of ZnO:Al were obtained by a hall measurement system with Van der Pauw method. The electron concentration and the mobility of ZnO:Al could be estimated from the Hall measurement results to be 1.2×10^{18} cm^{-3} and 3.4 cm^2 V^{-1} s^{-1}, respectively.

The comb-shaped Au Schottky electrode was made by using the photolithography technique to form Au/ZnMgO/ZnO:Al photodiode. The current–voltage (*I*–*V*) property and the time-dependent photocurrent were measured by a semiconductor analyzer (Agilent B1500A) at room temperature. A 150 W UV enhanced Xe lamp with a monochromator was used to investigate the spectral response property of the photodetector. The transient response spectra of the device were recorded by using an oscilloscope (Tektronix DPO 5104 digital oscilloscope) and a Nd:YAG laser (266 nm).

3. Results and discussion

Fig. 1(a) and (b) show the cross-sectional and surface SEM images of the ZnMgO/ZnO:Al on *c*-Al_2O_3 substrate, respectively. In Fig. 1(a), the thickness of the ZnO:Al layer and ZnMgO layer can be estimated to be 200 nm and 600 nm, respectively. Besides, it can be seen from Fig. 1(b) that the surface of the ZnMgO film is uniform and relatively smooth, which is further confirmed by the AFM image. As shown in Fig. 1(c), the surface of ZnMgO film is flat with a root-mean-square roughness of 1̃.11 nm. In order to investigate the structural properties, the XRD measurement was carried out for three typical samples: ZnO:Al, ZnMgO, and ZnMgO/ZnO:Al, as shown in Fig. 1(d). Besides the (0006) diffraction peak of the sapphire substrate, only one diffraction peak can be clearly observed for three samples. The diffraction peak located at around 34.6° correspond to the (0002) orientation of wurtzite ZnMgO and ZnO:Al. Apparently, after introducing Mg into ZnO, the (0002) diffraction peak moves to the large angle side because of the substitution of Zn atoms by Mg atoms. In addition, the full width at half maximum (FWHM) of the (0002) diffraction peak for ZnO:Al is relatively larger than that for ZnMgO, which could be explained by the poor crystal quality of ZnO deposited by RF magnetron sputtering than that of ZnMgO fabricated by MBE. As a result, it is difficult to distinguish

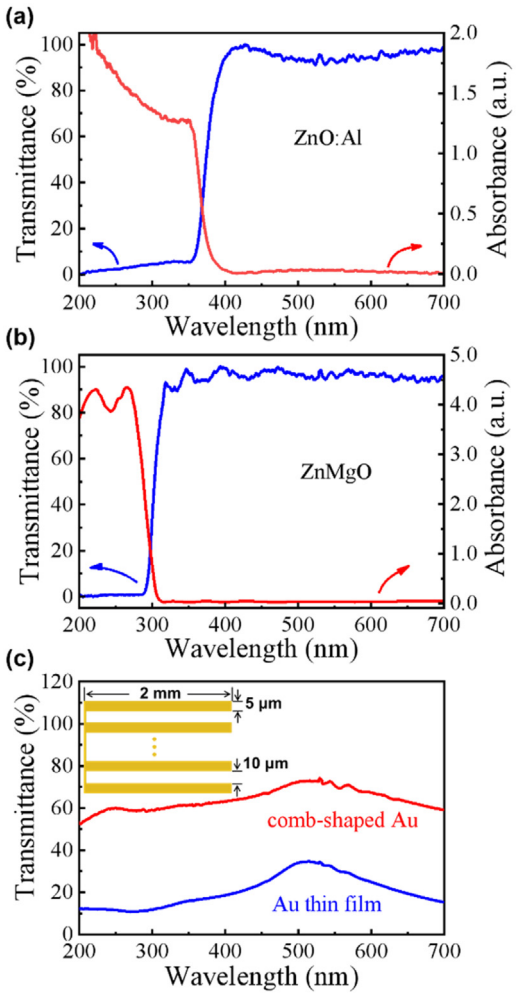

Fig. 2. Transmission and absorption spectra of the ZnO:Al film (a) and ZnMgO film (b) on the sapphire substrate. (c) Transmission spectra of Au thin film structure and comb structure. The inset is the schematic of comb structure.

the diffraction peaks of the ZnO and ZnMgO from the XRD spectrum of the ZnMgO/ZnO:Al on sapphire substrate as shown in Fig. 1(d). The composition of ZnMgO was determined to be $Zn_{0.58}Mg_{0.42}O$ by the EDS result in Fig. 1(e). Fig. 2(a) and (b) show the transmission and absorption spectra of the ZnO:Al and ZnMgO film on sapphire substrate, respectively. It is clear that the average transmittance of both two samples is more than 85% in the visible region. And the ZnO:Al and ZnMgO films have sharp absorption edges at ~360 nm and ~290 nm, respectively. Fig. 2(c) shows the transmission spectra of Au thin film structure and comb structure with a thickness of 30 nm. The schematic structure of comb-shaped Au is presented in the inset of Fig. 2(c), and the comb part of the structure is the Au micro-grating with the width of 5 μm, the length of 2 mm and a gap of 10 μm between the adjacent gratings. Obviously, the average transmittance of comb-shaped Au is more than 60% in the wavelength range between 200 nm and 300 nm, while this value is only around 10% for Au thin film. Therefore, the use of the comb-shaped Au as an electrode is expected to effectively improve the light absorption efficiency of the photosensitive layer in an UV photodetector.

The comb-shaped Au/ZnMgO/ZnO:Al vertical Schottky photodetector was demonstrated and investigated. As shown in the inset of the Fig. 3(a), com**b-**shaped Au and ZnO:Al layer were used as Schottky and ohmic electrodes of the ZnMgO, respectively. Fig. 3(a) is the *I–V* curve of the Au/ZnMgO/ZnO:Al photodetector in the dark at room temperature. And a weak rectification can be clearly observed in the dark condition, which may be associated with the defect-related tunneling effect at the Au/ZnMgO interface and the large series resistance of ZnMgO film [29,30]. Fig. 3(b) shows the spectral response of the device at zero bias. Obviously, the device displays a response peak at ~265 nm with a -3 dB cutoff wavelength of ~283 nm, and no obvious response peak at around 380 nm associated with the bottom ZnO:Al layer can be found, indicating that this device can be used for solar-blind UV photodetection without any external power supply. The repeatability is an important factor in determining the long-term stable dynamic operation of a photodetector. The time dependence of photocurrent property is measured at zero bias by switching ON/OFF 265 nm light illumination with different intensities (from 1.1 μW/cm^2 to 1.1 mW/cm^2) as shown in Fig. 4(a). It can be clearly observed that the device exhibits good ON/OFF switching performance with high stability and reproducibility. And the saturated photocurrent increases with increasing the light intensity. Fig. 4(b) describes the responsivity and photocurrent of Au/ZnMgO/ZnO:Al UV detector as a function of the UV light (265 nm) intensity. With decreasing the light intensity from 1.1 mW/cm^2 to 80 nW/cm^2, the responsivity of the device could increase from 2.58 mA/W to 55 mA/W, with the corresponding photocurrent decreasing from 14.2 nA to 0.022 nA. This phenomenon could be explained by the fact that a large number of photogenerated carriers are generated under strong light conditions, resulting in a high recombination rate of carriers. To the best of our knowledge, the peak responsivity of 55 mA/W for our Au/ZnMgO/ZnO:Al photodetector at zero bias is the highest value for the reported ZnO-based self-powered UV detectors [2,4,14,17,18,27,31–36]. The high responsivity of our device can

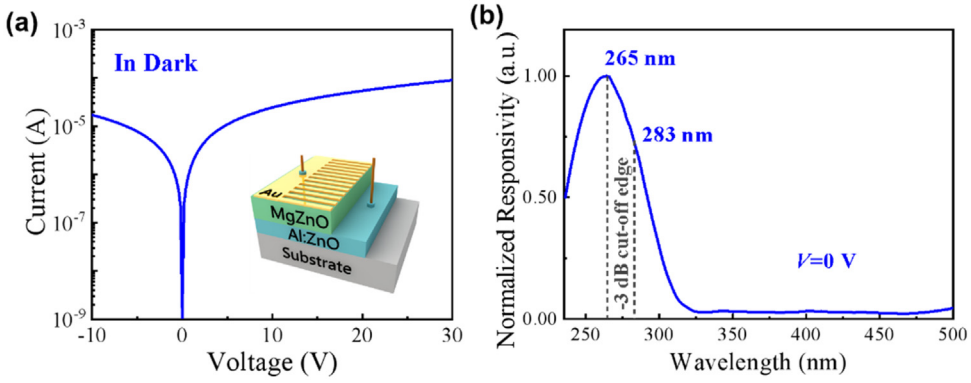

Fig. 3. (a) Current–voltage characteristics in the dark condition and (b) Spectral response property at 0 V of the Au/ZnMgO/ZnO:Al photodetector. The inset in (a) is the schematic illustration of the Au/ZnMgO/ZnO:Al vertical structure.

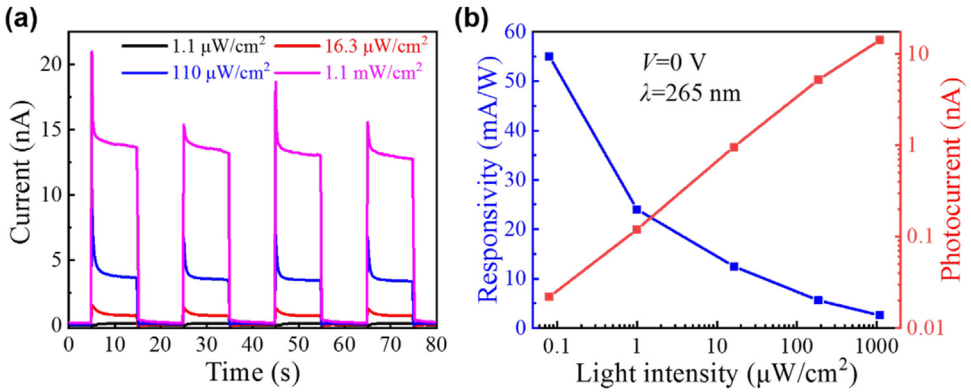

Fig. 4. (a) Time-dependent photocurrent of the device at zero bias under 265 nm light illumination with different intensities. (b) The responsivity and the photocurrent as a function of the 265 nm light intensity at 0 V.

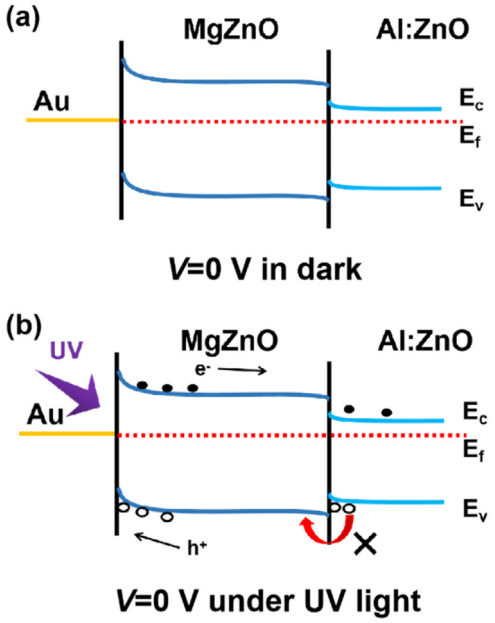

Fig. 5. Energy-band diagram of the comb-shaped Au/ZnMgO/ZnO:Al photodetector at 0 V in the dark (a) and under UV irradiation (b).

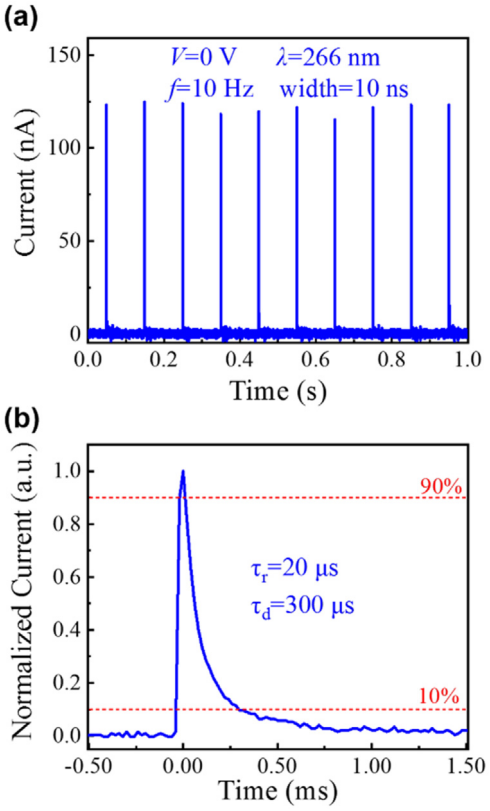

Fig. 6. (a) Transient photoresponse of photodetector at zero bias under the illumination by a 266 nm pulsed laser with a frequency of 10 Hz, (b) is the enlarged view of a single cycle.

be attributed to the high transmittance of the comb-shaped Au electrode.

Fig. 5(a) shows the band diagram for the Au/ZnMgO/ZnO:Al system. When the device is irradiated by the UV light, the photon with energy larger than bandgap of ZnMgO will be absorbed. Then electrons–hole pairs are photo-induced and the build-in electric field at the Au/ZnMgO interface drives the photo-generated electrons toward ZnO side, while photo-generated holes shift to the Au side. As a result, photocurrent signal can be obtained at 0 bias induced by solar-blind UV light. When the device is irradiated by the UV light with the photon energy between the bandgap of ZnMgO and ZnO:Al, the photon will be absorbed in ZnO:Al layer, inducing the photo-generated carriers. However, owing to the band offset (see Fig. 5(b)), these photo-generated carriers cannot contribute to the overall device photocurrent, which can be used to explain why our device has no obvious photoresponse to UVA.

To investigate the response speed of Au/ZnMgO/ZnO:Al photodetector, the transient response property was measured at 0 V by using a pulsed Nd:YAG laser with a wavelength of 266 nm (the laser pulse width was 10 ns, and the frequency was 10 Hz). As shown in Fig. 6(a), the photoresponse of our Au/ZnMgO/ZnO:Al photodetector is very fast, highly stable and reproducible. The rise time t_r and decay time t_d (defined as the time interval of the current rising from 10% to 90% and dropping from 90% to 10% of the peak values) are only 20 μs and 300 μs, respectively (see Fig. 6(b)).

Table 1 summarizes the typical responsivity and response time of the reported ZnO-based self-powered photodetectors. It is clear that our comb-shaped-Au/ZnMgO/ZnO:Al photodetector has the largest responsivity at 0 V. Additionally, the response speed of our device (t_r = 20 μs, t_d = 300 μs) is much faster than that of most reported devices. Although a rise time of 10 ns and a decay time of 150 ns have been observed for ZnMgO p-n junction photodetector, its responsivity was only 3.7×10^{-3} mA/W at zero bias [32].

Table 1
Peak responsivity at zero bias and response time of typical ZnO-based self-powered UV photodetectors.

Photodetector	Peak responsivity at 0 V	Rise time	Decay time	Ref.
PANI/ZnMgO	0.16 mA/W @250 nm	300 ms	300 ms	[2]
Ag/ZnMgO/ZnO	16 mA/W @275 nm	0.024 ms	0.3 ms	[4]
ZnMgO:Al/PEDOT:PSS	19.1 mA/W @278 nm	320 ms	200 ms	[14]
ZnO MSM	20 mA/W @365 nm	0.00071 ms	0.004 ms	[17]
ZnMgO:Sb/n-Si	1.57 mA/W @320 nm	100 ms	100 ms	[18]
Pt/ZnMgO	15 mA/W @220 nm	–	–	[27]
Ga2O3/ZnO:Ga	0.76 mA/W @260 nm	179 ms	272 ms	[31]
n-ZnMgO/p-ZnMgO	3.7×10^{-3} mA/W @325 nm	0.00001 ms	0.00015 ms	[32]
p-ZnO:(Li,N)/n-ZnO	0.018 mA/W @380 nm	–	–	[33]
Se/ZnO	2.65 mA/W @370 nm	0.69 ms	13.5 ms	[34]
CuZnS/ZnO	12 mA/W @300 nm	700 ms	860 ms	[35]
p-GaN/n-ZnO	0.68 mA/W @358 nm	–	–	[36]
Au/ZnMgO/ZnO:Al	55 mA/W @265 nm	0.02 ms	0.3 ms	This work

4. Conclusions

In summary, a self-powered solar-blind UV photodetector was realized based on Au/ZnMgO/ZnO:Al vertical structure on the *c*-sapphire substrate with comb-shaped Schottky electrode. The ZnO:Al layer was used both as a buffer layer for growing ZnMgO layer and as a bottom ohmic electrode for the photodetector in this work. The Au Schottky electrode with comb-shaped structure shows the average transmittance of more than 60% in the wavelength range between 200 nm and 300 nm. At 0 V bias, the Au/ZnMgO/ZnO:Al detector shows a response peak at 265 nm with a -3 dB cut-off wavelength of 283 nm, indicating that it can be used for solar-blind detection. And the responsivity for a weak UV light illumination (265 nm, 80 nW/cm^2) could reach as high as 55 mA/W, which is larger than that of any other reported ZnO-based UV photodetectors at 0 V. Moreover, a fast response speed can be clearly observed with 10%–90% rise and 90%-10% decay times of 20 μs and 300 μs, respectively. This work provides a viable way to achieve high performance ZnO-based solar-blind UV detectors for weak signal detection.

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