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Flexible, transparent and self-powered deep ultraviolet photodetector based on Ag NWs/amorphous gallium oxide Schottky junction for wearable devices

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ABSTRACT

Wearable photodetector, owing to the significant development of intelligent and integrated science and technologies, have caused great concern recently on account of their wide potential applications in smart monitoring. Self-powered is the trend of future development for the wearable photodetector due to the increased demand of energy-saving, miniaturization and high efficiency in devices. Gallium oxide, with a bandgap energy of 4.9 eV, is a perfect material for the next generation deep ultraviolet (DUV) photodetectors. In this work, Ag nanowires (NWs)/amorphous gallium oxide (a-GaO) Schottky junction was constructed on the conductive flexible substrate for self-powered DUV wearable photodetectors. The photoelectricity performance of Ag NWs/a-GaO based DUV photodetector is related to the density of Ag NWs electrode. Under the zero bias, the photodetector shows a photoresponse time of 0.24 s, a high light-to-dark ratio of 120 and a good repeatability under 254 nm light illumination with a light intensity of 3000 $\mu W/cm^2$. Moreover, it exhibits the excellent mechanical properties, whose photoelectric performance is almost unchanged under the strain of 0.25 %. The Ag NWs/a-GaO based DUV photodetector combining the flexible with self-powered property, meets great well with the wearable device requirements.

1. Introduction

Wearable photodetector, owing to the significant development of intelligent and integrated science and technologies, have caused great concern recently on account of their wide potential applications in smart monitoring [1,2]. Research on wearable photodetector becomes a hot topic worldwide in the past decade and the number of publications on wearable photodetector exhibits the exponential growth as a function of time on logarithmic scale. Compared with traditional photodetectors, which should satisfy the "5S" requirements, wearable devices need more features, such as keep high performance and good stability under bending, compressing or twisting [3]. In addition, in order to realize the functions of wearable photodetector, some additional units such as power supply, data storage and display are usually required. With the rapid development of integrated system, the wearable photodetector are gradually developing towards self-powered because the traditional wearable power supplies are unable to meet the need of complex integrated system [4,5].

In order to construct high performance wearable photodetector, careful selection of light absorption materials of photodetector should be given priority. Ga_2O_3 semiconductor has a large bandgap energy of 4.9 eV, is a natural light absorbing material for DUV photodetectors [6–10]. In recent years, Ga_2O_3 based solar blind photodetectors gradually become a research hotspot on account of the advantages of high spectral selectivity and low false alarm rate, which is crucial to civilian and military fields [9]. To meet the requirements of wearable device, gallium oxide based photodetectors desire to be flexible and self-powered. However, most reported high crystalline quality gallium oxide based photodetectors need to be prepared on a substrate at high temperature (>800 °C), while soft substrates generally cannot stay flexible at this temperature [11,12]. Fortunately, owing to the low growth temper-

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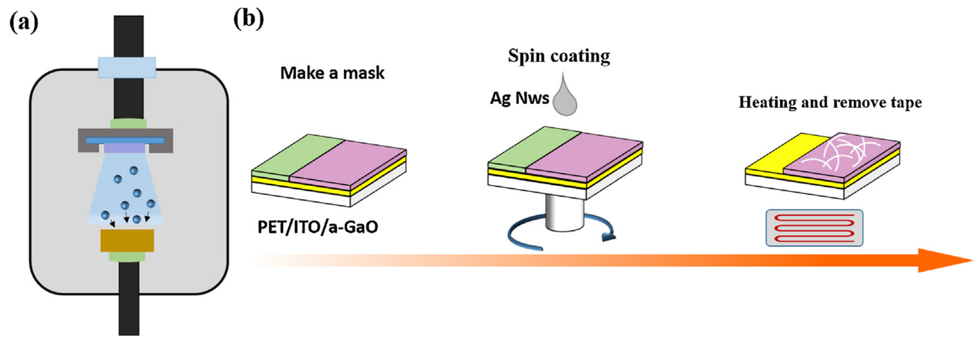

Fig. 1. Scheme of fabrication process of Ag NWs/a-GaO Photodetector.

ature and high optoelectronic performance of amorphous gallium oxide (a-GaO), gallium oxide based flexible photodetector have been realized [13–18]. Based on a-GaO films, a flexible photodetector has been fabricated on polyethylene naphthalate substrate at room temperature, which shows a high photocurrent-to dark current ratio (PDCR,$>10^4$) and a response speed (<20 μs) [19]. A visible-NIR transparent photodetector, constructed of a-GaO and amorphous In-*Zn*-O electrode at room temperature, demonstrates an ultrahigh photoresponsivity and external quantum efficiency under a positive bias [19]. However, they must work under external power supply, which is not suitable for a smart wearable devices. Some new novel constructions and techniques are need to further realize Ga_2O_3 based wearable device.

Recently, photodetectors based on photovoltaic effect, which origin from pn junction, heterojunction or Schottky junctions, have been developed rapidly, which can work without power supply. After years of development, the photoelectric performance of photodetectors can satisfy the human demands, but the complex and expensive preparation process of p-n junctions and heterojunction limit the industrial production of self-powered photodetector [20–29]. Theoretically, Schottky junctions based on metal-semiconductor contact is an ideal device structure for commercial gallium oxide self-powered photodetectors owing to its low turn-on voltage, simple and economical preparation process. As reported in previous literatures, the low transmission of the traditional film type upper electrode is the drawback for Schottky-type photodetector [28]. Excitingly, after years of development and improvement, a series of transparent electrodes, such as ITO, graphene and silver nanowires (Ag NWs), has emerged as promising upper electrode for developing efficient Schottky-type devices. However, the low transmittance of ITO in the DUV region limits its application in DUV photodetector. Meanwhile, complex growth and transfer processes limit the application of graphene electrodes in industrial production. Among them, the Ag NWs becomes a preferential upper electrode for its outstanding properties, such as high transmittance and electric conductivity, low cost and easy fabrication process.

In this research, a large-detective-area šandwichštructure self-powered flexible photodetector based on Ag NWs/a-GaO film Schottky junction was successfully completed on terephthalate/indium tin oxide (PET/ITO) substrate at room temperature. The photogenerated electron–hole pairs in the space charge can be spontaneous separate, hence, the Schottky junction photodetectors exhibit self-powered property. Under zero bias, the photodetector shows a rise speed of 0.24 s, a high PDCR of 120 and a good repeatability under the irradiation of a wavelength with 254 nm. Moreover, it exhibits the excellent mechanical properties, whose photoelectric performance is almost unchanged under the strain of 0.25 %. The Ag NWs/a-GaO based DUV photodetector combining the flexible with self-powered property, meets great well with the wearable device requirements.

2. Method

2.1. Preparation of the Ag NWs/a-GaO/ITO/PET photodetector

The ITO coated PET films were purchased from Zhuhai Kaiov Optoelectronic Technology Co., LTD. The sheet resistance of the ITO/PET film is 7 O/sq. Ag NWs alcoholic solution was obtained from Alfa Aesar Co., Ltd. The concentraion of Ag NWs alcoholic solution is 5 mg/mL. The Ag NWs used here possess a diameter about 150 nm and a length of 10–100 micrometers. A series of a-GaO films were deposited on ITO/PET substrates by magnetron sputtering at room temperature. The radio frequency power and Ar flow rate was 200 W and 10 sccm, respectively. The deposition time was set as 0.5 h, 1 h, 2 h and 3 h. The Ag NWs alcohol concentration is diluted to 0.4 mg/mL. Ag NWs was spun at 200 r/s for 5 s and 5000 r/s for 10 s. The sample coated with Ag NWs was baked at 120 °C for a quarter hour. Ti/Au electrodes were deposited on the edge of the Ag NWs film and a-GaO film to accomplish the fabrication of the detector. The effective illumination area is 1 cm^2.

2.2. Characterization of the materials and the photodetector

The morphologies of Ag NWs and the cross profile of as-grown a-GaO film were characterised by a SEM. The transmittance of Ag NWs and a-GaO were measured by UV − vis spectrophotometer (Hitachi U-3900). Transmittance of Ag NWs and photodetector were test by a UV–vis spectrophotometer (U-3900). Photoelectric performance ($I - V$ and $I - T$ characteristic curves) of the photodetector was measured by Keithley 4200.

3. Results and discussion

The fabrication process is schematically illustrated in Fig. 1. Firstly, ITO/PET substrates were wiped with alcohol and dry under nitrogen. Then, a series of a-GaO films with different growth time were prepared by magnetron sputtering at room temperature. Magnetron sputtering technique is one of the well-established physical vapor deposition techniques used in industry and research due to its advantages such as low cost, low deposition temperature, high reproducibility and scalability [30–32]. Thirdly, a uniform Ag NWs electrode were formed on as-grown a-GaO film by spin-coating process and baked at 120 °C for a quarter hour. After drying, the Ag NWs spontaneously form a two-dimensional conducting channel. Finally, the photodetector was successfully prepared by depositing Ti/Au electrode. Fig. 2a shows the structure diagram of the Ag NWs/a-GaO device, in which the a-GaO film is situated between Ag NWs and ITO electrodes forming a sandwich structure. This sandwich structure is helpful to photogenerated carriers separate, which enhance the effect of built-in electric field.

The top view field emission scanning electron microscope (FESEM) image of the Ag NWs fabricated on a-GaO film as the

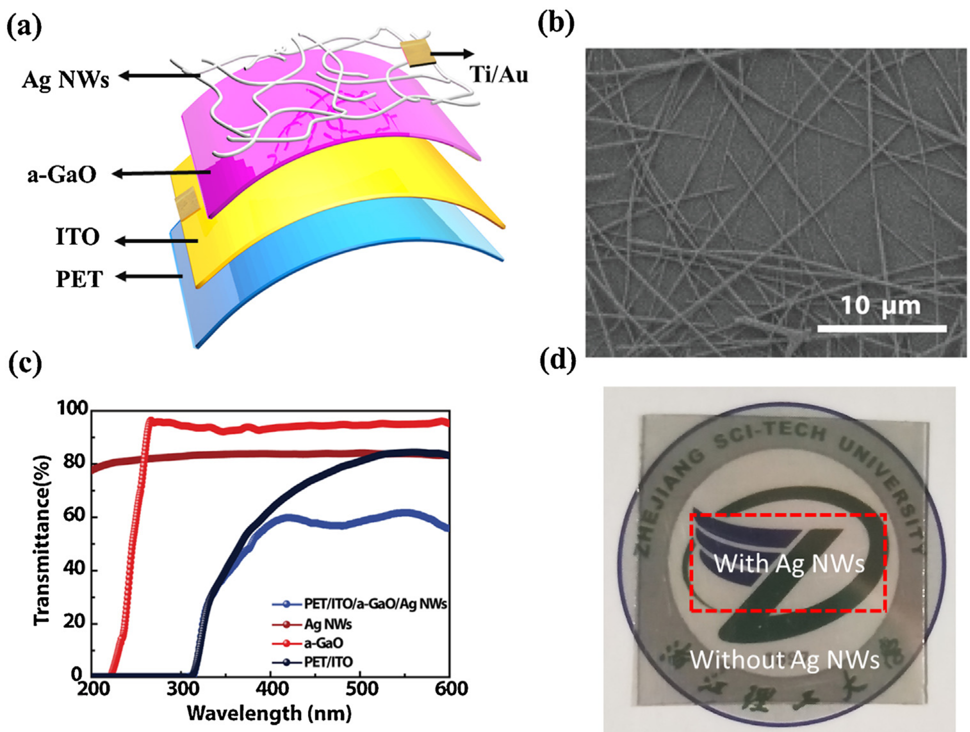

Fig. 2. (a) Schematic illustration of the photodetector; (b) SEM image of Ag NWs electrode; (c) Transmittance spectrum of the ITO/PET substrate, Ag NWs, and the photodetector; (d) Digital image of the transparent solar - blind photodetector.

top electrodes was presented in the Fig. 2b. The Ag NWs used here is possess a diameter about 150 nm and a length of 10–100 micrometers. Meanwhile, individual nanowires connect smoothly and tightly mutually, providing a two-dimensional conducting channel. Both Ag NWs electrode and a-GaO film are transparent in visible region which show a high transmittance above 80 %, as shown in Fig. 2c. The sharp drop transmittance of the device in DUV region is caused by the absorption of UV light by ITO layer and PET substrate. The transparency of the device is further confirmed by the photograph in the Fig. 2d.

The upper electrode of the Ag NWs is crucial to the Ag NWs/a-GaO based photodetector. In this work, to optimize the Ag NWs electrode, Ag NWs with a various density were obtained by repeating the above process. Fig. 3a shows the transmittances of the Ag NWs electrodes as a function of the cycle number of spin-coating. The transmittance of the Ag NWs electrode decreases monotonically as the cycle number increases. With the cycle number less than twenty, the Ag NWs electrode shows a high transmittance of above 80 % in the DUV region. To further investigate the electrical conductivity of the Ag NWs, photoelectric performance of the a-GaO based photodetector with the different Ag NWs density electrodes were studied. With a bias of 0 V, the 254 nm UV illumination was turned on and off intermittently to measure the time-dependent photoresponse of the device. With the increase of the Ag NWs density, the photocurrent of the device first increase then decrease and achieve the maximum value of 12.05 nA with the cycle number of twenty (Fig. 3b). The increase of the photocurrent is origin to the eliminated of open space between non-connected Ag NWs and the optimized Ag NWs electrode with the cycle number of twenty is tradeoff between conductivity and transmittance.

The light absorption layer of the a-GaO thin film is another important factor in our Ag NWs/a-GaO based self-powered DUV photodetector. The photocurrent of the photodetector first increases then decreases as the deposition time of the a-GaO thin film increases, which reaches the maximum value of 12.05 nA with 2 h (Fig. 3c). As the thickness of the a-GaO thin film increases, the photogenerated carriers increase, owing to the increase of the incident absorption light. However, as the thickness continues to increase, some carriers have been recombined before being collected by the electrode. As a result, the Ag NWs/a-GaO based photodetector with the a-GaO thin film deposited 2 h exhibits the highest photocurrent. All the following studies are based on the optimized conditions described above. SFig. 1 shows the X-ray diffraction pattern of the sputtered a-GaO film on Al_2O_3 substrate for 2 h at room temperature. There is only Al_2O_3-related diffraction peaks, which indicate that the obtained gallium oxide film is amorphous. Fig. 3d and e present the thickness and absorption spectrum of the a-GaO film. The uniform 460 nm a-GaO film can be clearly observed in the cross-sectional SEM morphology. From the UV–vis absorption spectrum, it is clear that a-GaO film has a significant absorption at DUV region (<280 nm), corresponding to a band gap of ~4.72 eV (the inset of Fig. 3d). To test the DUV detective ability of Ag NWs/a-GaO photodetector, photoelectric performance of the photodetector were mearsured. The current-voltage (*I-V*) curves of the device are nonlinear and asymmetric (Fig. 4a), which displays an off-state current at the reverse bias and an on-state current at the forward bias, exhibiting a rectification characteristic. In order to fully explain the mechanism of the photodetector, energy band diagrams of the device is proposed, as shown in Fig. 4b. It was reported that ITO contacts on Ga_2O_3 showed an Ohmic contact. And the work function of Ag (4.6 eV) is bigger than electron affinity of a-GaO (4.0 eV) [33,34]. Hence, Schottky junction is formed between a-GaO film and Ag NWs electrode (as illustrated in Fig. 4b), and photoinduced electron-hole pairs can be separated effectively. The Schottky barrier of the Ag-GaO contact, which can be described by the thermionic emission-based diode equation:

$$J(T, V) = J_s(T)\left[exp\left(\frac{eV}{\eta K_B T} - 1\right)\right]$$

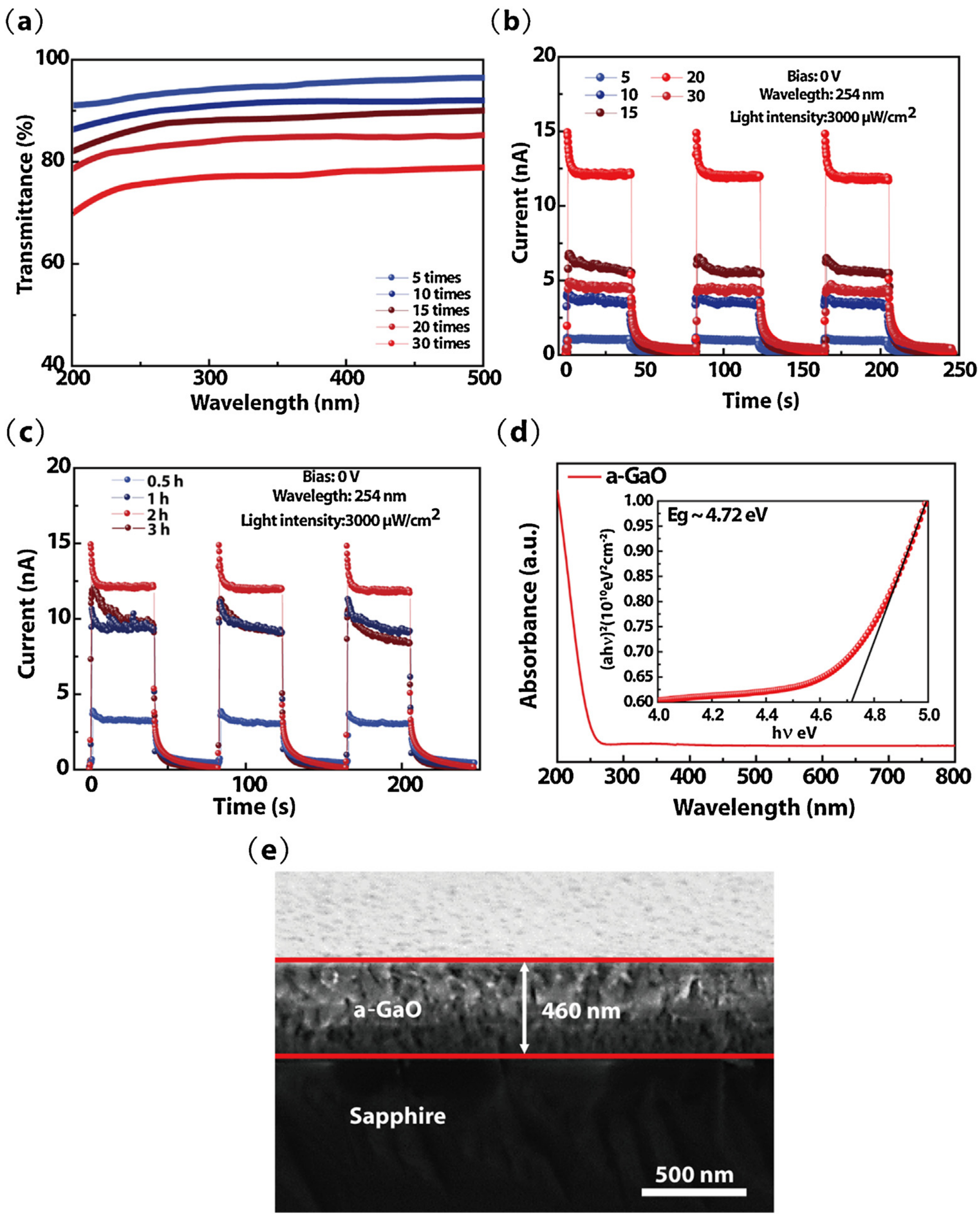

Fig. 3. (a) Transmittance spectrum of the Ag NWs with different cycle numbers; (b) Time-dependent photoresponse of the Ag NWs/a-GaO photodetector with different density Ag NWs top electrodes under zero bias and 254 nm light; (c) Time-dependent photoresponse of the photodetector with different deposition time a-GaO film; (d) The cross-section of a-GaO film, (e) Optical absorbance spectra of a-GaO film (the inset is the band gap of the a-GaO film).

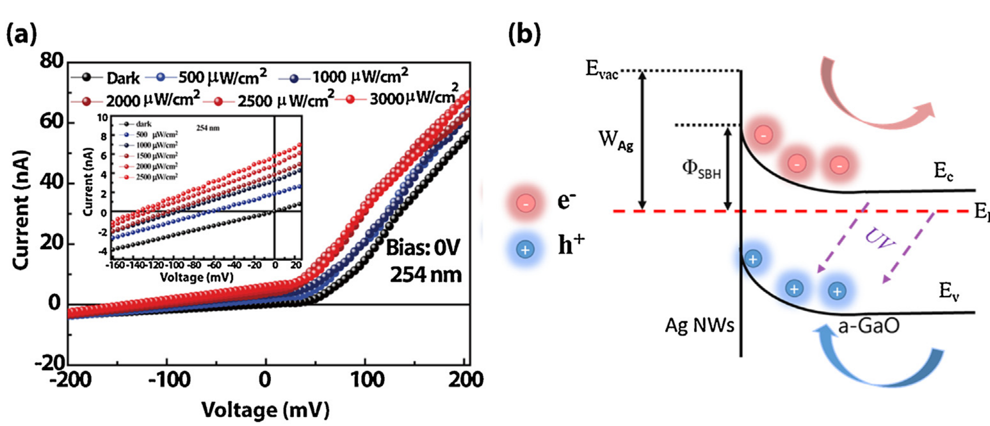

Fig. 4. (a) $I-V$ characteristic curve of the Ag NWs/a-GaO based photodetector under dark and 254 nm illumination with various light intensities, the inset is the $I-V$ enlarged curves near zero bias; (b) Energy band diagram of the device.

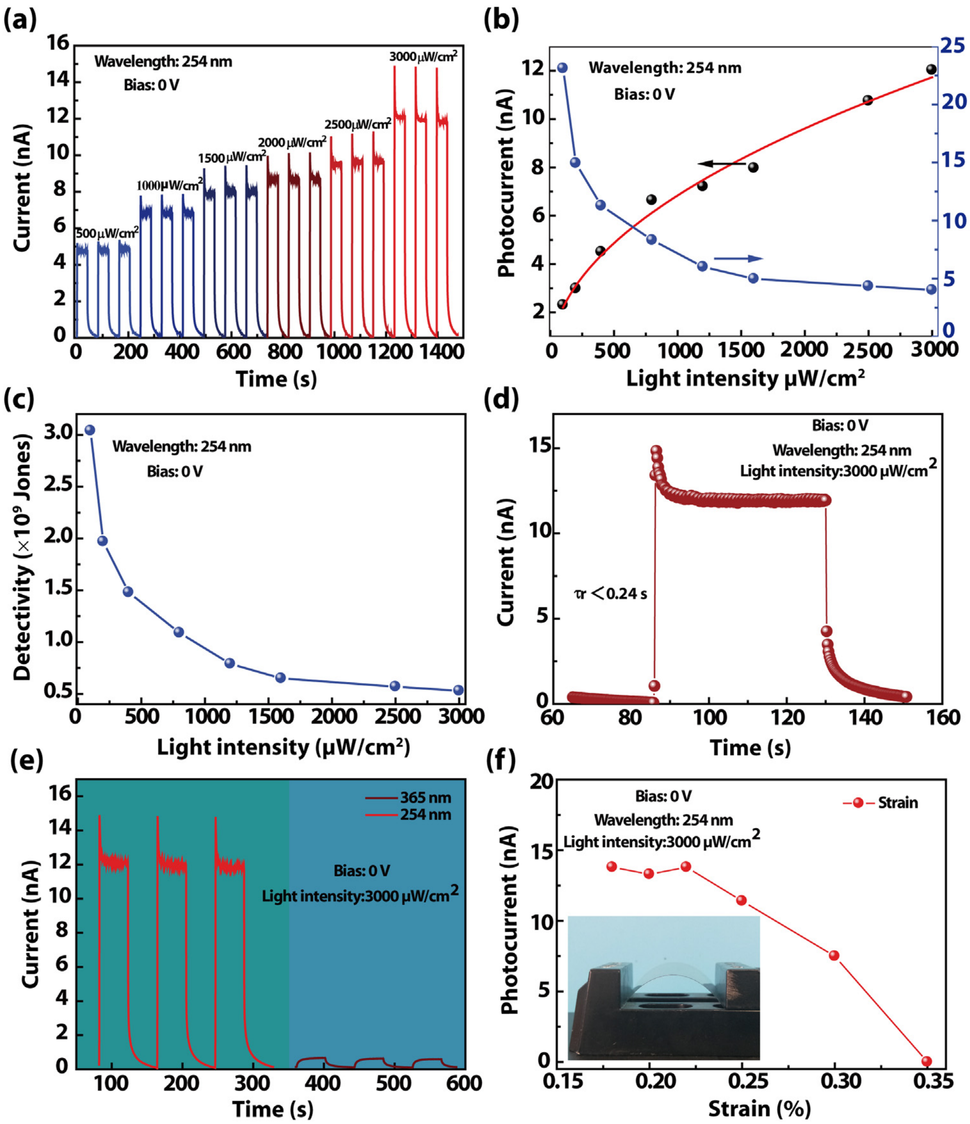

Fig. 5. (a) Time-dependent photoresponse of the photodetector under zero bias and 254 nm light with various light intensities; (b) Photocurrent and responsivity as a function of the light intensity; (c) Detectivity as a function of the light intensity; (d) Enlarged view of the rise/decay edges; (e) Time-dependent photoresponse of the Ag NWs/a-GaO junction photodetector under zero bias at 254 and 365 nm illumination; (f) Variations in photocurrent of the device under 254 nm UV light illumination with different strains.

where J(T, V) is the current density across the Ag-GaO interface, V is the applied voltage, k_B is the Boltzmann's constant, T is the absolute temperature, and n is the ideality factor. The prefactor, Js(T) is the saturation current density and can be described by $J_s(T) = A^*T^2e^{\Phi_{SBH}/K_BT}$, where Φ_{SBH} is the zero bias Schottky barrier height (SBH), A* is the Richardson constant. For GaO material, A* is theoretically calculated to be 41.1A cm^{-2} k^{-2} ; using the Js value, the Schottky barrier height at the Ag-GaO interface is estimated to be 0.8 eV [35,36].

To study systematically, *I-t* measurements were performed by switching on and off 254 nm light regularly. The light intensity of the 254 nm light is range from 500 to 3000 μW/cm^2 (Fig. 5a). At 0 V, the current is 0.10 nA in dark and increases to 12.05 nA under 3000 μW/cm^2 intensity 254 nm light, corresponding to a PDCR of 120. The results from above show that the Ag NWs/a-GaO photodetector is a zero energy consumption device. As the light intensity increase from 500 to 3000 μW/cm^2, the photocurrent increases linearly from 4.69 nA to 12.05 nA, which is because of more photogenerated electron-hole pairs produced by light absorption layer under a higher intensity light. The functional relationship between the photocurrent and light intensity is plotte in Fig. 5b, which can be well fitted by the Equation:

$$I_{ph} \propto P^{\gamma} \tag{1}$$

here, *P* is the light intensity and γ is a factor. The γ is calculated to be 0.52 for Ag NWs/a-GaO film solar blind photodetector at 0 V under 254 nm light. Such a nonunity exponent (< 1) is owing to the recombination and trapping of electron-hole pairs by oxygen vacancy traps in the a-GaO film. The sensitivity of photodetector was evaluate by calculating responsivity (*R*) by the following formula: $R = (I_{photo} - I_{dark})/(PS)$, where I_{photo} and I_{dark} is the photocurrent and dark current, respectively, *P* is the supplied light intensity, and *S* is the effective area of the photodetector. As shown in Fig. 5b, under zero bias and 254 nm light illumination, the responsivity of the photodetector decreases as the light intensity increase and achieve the maximum value of 0.23 μA /W at 100 μW/cm^2. Though, more electron-hole pairs will be generated at a higher intensity light, the recombination also increase. The detection of the photodetector was evaluate by calculating detectivity (*D*). As shown in Fig. 5c, the *D* decreases as the light intensity increases. The maximum D

Table 1
The device parameters comparison of some a-GaO based photodetectors.

Photodetector	I_{photo}/I_{dark}	Response time	Flexibility	Self-powered	Ref
ITO/a-GaO/ITO	$>10^4$ (10 V)	19.1 (μs)	YES	NO	21
Ti/Al/a-GaO/Ti/Al	$>10^3$ (10 V)	0.41 (s)	NO	NO	25
Ag/β-Ga_2O_3/fiber	~260 (20 V)	0.4 (s)	YES	NO	26
IZO/a-GaO/IZO	~100 (20 V)	6.1 (s)	YES	NO	27
Pt/a-GaO/Pt	~1000 (20 V)	3 (μs)	NO	NO	12
Ag NWs/a-GaO/ITO	~120 (0 V)	<0.24 (s)	YES	YES	This work

value of this photodetector is about 3.1 × 10^9 Jones. For a more detailed research, the photoresponse time is fitted to calculate the response time [37–39]. The fitting results of the rise time are given in Fig. 5d, which the rise time (t_r) is defined as the time increasing from 10 % to 90 % of the maximum of photocurrent. The rise time of Ag NWs/a-GaO photodetector is about 0.24 s under 254 nm light illumination indicating that the construction of the Schottky junction effectively promotes the response speed [40–42]. The spectral selectivity of the photodetector was verified by measured the photoelectric performanceof the Ag NWs/a-GaO photodetector under 365 nm light radiation. The rejection ratio (R_{254}/R_{365}) of the junction photodetector at 0 V is about 70, which presents a satisfactory spectral selectivity of the light (Fig. 5e).

The mechanical properties are widely accepted to be vital to the reliability of flexible opotoelectronic devices. As shown in Fig. 5f, the photoelectricity of the Ag NWs/a-GaO photodetector under various bending conditions were measured. The varied strains are applied to the device by bending the PET substrate to different bending radius of curvature. Then the strain value was estimated through the following equation (SFigure. S3),

$$\text{Strain}\,(\%) = \frac{t_s + t_f}{2R_C} \tag{2}$$

where t_s and t_f were the thickness of ITO/PET substrate and a-GaO film, respectively; and R_c represented the bending radius of curvature. It is clearly shown that the as -fabricated photodetector was able to bear an ultimate strain up to 0.3 %. And the photocurrent was almost unchanged under the strain from 0 to 0.25 %. These results demonstrate the excellent mechanical flexibility and the potential application of the Ag NWs/a-GaO photodetector in the flexible device.

Table 1 compared the Ag NWs/a-GaO photodetector with previously reported a-GaO based photodetectors [13,22,26–28]. As an a-GaO based photodetector, the device shows a considerable photoelectric performance. More importantly, the a-GaO film based solar blind photodetector combining the flexible with self-powered property, which has been reported for the first time, meets great well with the wearable device requirements.

4. Conclusion

In summary, a large-detective-area šandwichštructure self-powered flexible photodetector based on a-GaO film was successfully completed on PET/ITO substrate at room temperature. Combining deposited a-GaO film process with solution-processed of Ag NWs electrode, the device fabricated by a simplicity, low cost and easy method. The photon-generated electron–hole pairs in the space charge can be spontaneous separate, hence, the Schottky junction photodetectors exhibit self-powered properties. Under the zero bias, the photodetector shows a rise speed of 0.24 s, a high PDCR of 120 and a good repeatability under the irradiation of a wavelength with 254 nm. Moreover, photoelectric properties of the device were almost unchanged under the strain of 0.25 %. This self-powered flexible Ag NWs/a-GaO solar blind photodetector meets great well with the wearable device requirements and has potential application in smart monitoring.

Author statement

Shunli Wang: Conceptualization, Investigation. Chao Wu: Investigation, Aiping Liu and Nie Zhao Writing- Original draft preparation. Fengmin Wu: Supervision. Fabi Zhang and Daoyou Guo: Writing- Reviewing and Editing.

Declaration of Competing Interest

The authors declare no conflict of interest.

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