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Photoelectric properties of β-Ga_2O_3 thin films annealed at different conditions

Tuo Sheng, Xing-Zhao Liu*, Ling-Xuan Qian, Bo Xu, Yi-Yu Zhang

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Abstract In this work, metal–semiconductor–metal solar-blind ultraviolet photoconductors were fabricated based on the β-Ga_2O_3 thin films which were grown on the *c*-plane sapphire substrates by molecular beam epitaxy. Then, the effects of β-Ga_2O_3 annealing on both its material characteristics and the device photoconductivity were studied. The β-Ga_2O_3 thin films were annealed at 800, 900, 1000, and 1100 °C, respectively. Moreover, the annealing time was fixed at 2 h, and the annealing ambients were oxygen, nitrogen, and vacuum (4.9×10^{-4} Pa), respectively. The crystalline quality and texture of the β-Ga_2O_3 thin films before and after annealing were investigated by X-ray diffraction (XRD), showing that higher annealing temperature can result in a weaker intensity of ($\bar{4}02$) diffraction peak and a lower device photoresponsivity. Furthermore, the vacuum-annealed sample exhibits the highest photoresponsivity compared with the oxygen- and nitrogen-annealed samples at the same annealing temperature. In addition, the persistent photoconductivity effect is effectively restrained in the oxygen-annealed sample even with the lowest photoresponsivity.

Keywords Molecular beam epitaxy; β-Ga_2O_3 thin films; Annealing; Photoconductor

T. Sheng, X.-Z. Liu*, L.-X. Qian, B. Xu, Y.-Y. Zhang
School of Microelectronics and Solid-State Electronics, University of Electronic Science and Technology of China, Chengdu 610054, China
e-mail: xzliu@uestc.edu.cn

X.-Z. Liu
State Key Laboratory of Electronic Thin Films and Integrated Devices, University of Electronic Science and Technology of China, Chengdu 610054, China

1 Introduction

Nowadays, solar-blind ultraviolet photoconductors can be used in wide application fields, such as flame detection, ultraviolet leakage inspection, and combustion process monitoring. Moreover, β-Ga_2O_3 has been considered as one of the most promising materials for the fabrication of solar-blind deep ultraviolet photodetector due to its wide bandgap energy of 4.8 eV, high saturation velocity, good thermal stability, and large mechanical strength [1–4]. Recently, it was reported that the β-Ga_2O_3 thin film grown on *c*-plane sapphire substrate by molecular beam epitaxy (MBE) can be adopted for the fabrication of metal–semiconductor–metal (MSM) solar-blind ultraviolet photoconductors [5, 6]. However, the device performance was degraded by the lattice mismatching between β-Ga_2O_3 thin film and sapphire substrate. It was reported that annealing may improve the quality of β-Ga_2O_3 thin film and accordingly the device performance [7, 8]. In this study, the β-Ga_2O_3 thin films with different annealing conditions were monitored by X-ray diffraction (XRD). Then, all the films were fabricated into MSM solar-blind ultraviolet photoconductors to investigate both current-voltage (*I*–*V*) characteristics and transient photoresponse. Finally, the annealing effects on photoelectrical properties of β-Ga_2O_3 thin film were discussed.

2 Experimental

Firstly, the β-Ga_2O_3 thin films with a thickness of 100 nm were grown on *c*-plane sapphire substrates by MBE with a base pressure of 6.6×10^{-6} Pa and a substrate temperature of 660 °C. On the one hand, gallium with a purity of 99.99999 % as the source of Ga was evaporated from an effusion cell, and the temperature of effusion cell was fixed at 940 °C. On the other hand, oxygen with a purity of 99.999 % as the source of O was sprayed via a radio-frequency (RF) plasma source, and the flow rate of oxygen gas and the input RF power of plasma source were maintained at 1 ml·min^{-1} and 300 W, respectively. Then, the β-Ga_2O_3 thin films were annealed at 800, 900, 1000, and 1100 °C, respectively. Moreover, the annealing time was fixed for 2 h, and the annealing ambients were oxygen, nitrogen, and vacuum (4.9×10^{-4} Pa), respectively. The crystalline quality and texture of the β-Ga_2O_3 thin films before and after annealing were tested through a high-resolution XRD system (Bede-D1) with a Cu Kα line ($\lambda = 0.1542$ nm) as the radiation source. Finally, an interdigital Ti/Al (20 nm/20 nm) electrode with an area of 0.024 mm^2 was deposited on each β-Ga_2O_3 thin film by conventional electron beam evaporation and lift-off process, and accordingly, a photoconductor was fabricated. Then, a rapid thermal annealing was carried out at 500 °C for 5 min in nitrogen ambient in order to reduce the contact resistance. The electrode fingers were 3 μm in width, 80 μm in length, and 3 μm in spacing gap. In order to measure the *I*–*V* characteristics and transient photoresponse of the fabricated devices, a HP-4155B semiconductor parameter analyzer was utilized. Moreover, an ultraviolet lamp with a wavelength of 254 nm, combined with a time shutter, acted as the light source, whose illumination intensity was 85 μW·cm^{-2}. In addition, the measurement of transient photoresponse was taken at a constant voltage of 20 V.

3 Results and discussion

Figure 1 shows XRD patterns of β-Ga_2O_3 thin films annealed at different conditions. Figure 1a–c shows that the peak location of diffraction is almost unchanged for each annealing condition, indicating that annealing has no significant effect on the crystal orientation of β-Ga_2O_3 film. Moreover, there are four obvious peaks in each XRD pattern. Among them, the peak located at 41.7° is corresponding to (0006) of the sapphire substrate, and the peaks located at 18.9°, 38.4°, and 59.1° are corresponding to $(\bar{2}01)$, $(\bar{4}02)$, and $(\bar{6}03)$ of β-Ga_2O_3 thin film, respectively. Furthermore, the intensity of each diffraction peak was normalized based on the $(\bar{2}01)$ diffraction peak. In addition, Fig. 1d shows that the relative intensity of $(\bar{4}02)$ diffraction peak reduces with the increase in annealing temperature for each annealing ambient, which is possibly attributed to recrystallization of β-Ga_2O_3 thin film during

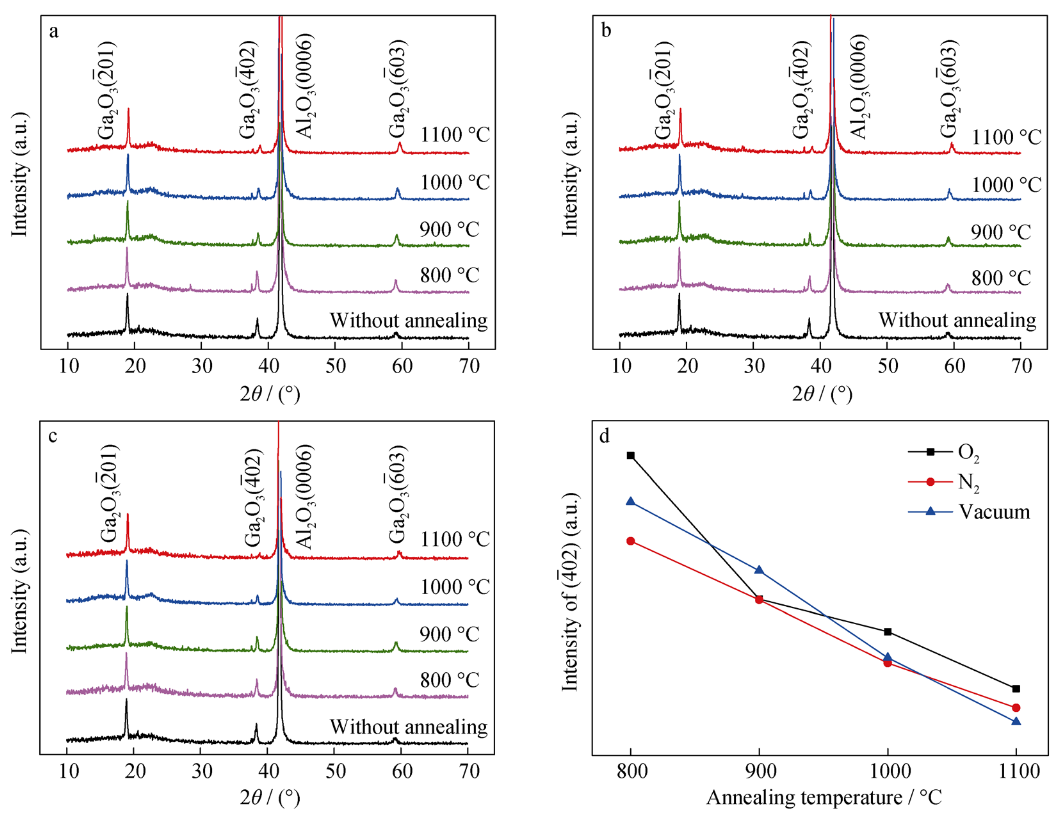

Fig. 1 XRD results of β-Ga_2O_3 thin films before and after annealing at different conditions: **a** annealing in oxygen, **b** annealing in nitrogen, **c** annealing in vacuum (4.9×10^{-4} Pa), and **d** correlation between $(\bar{4}02)$ intensity and annealing temperature

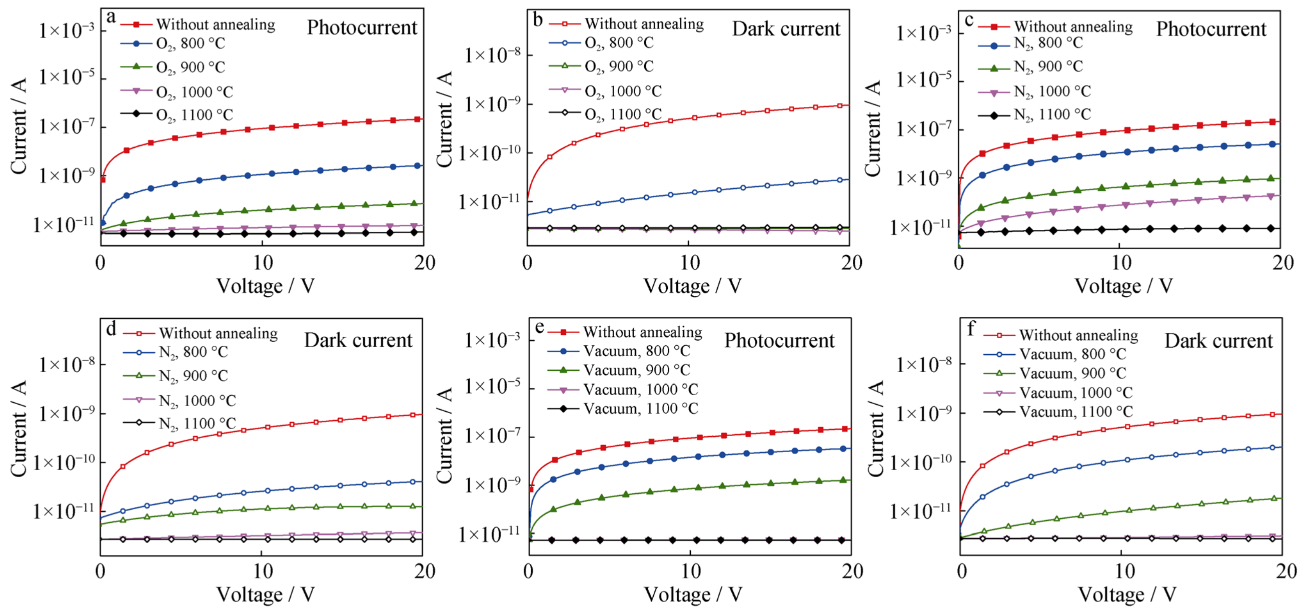

Fig. 2 *I–V* characteristics of as β-Ga_2O_3 photoconductors before and after annealing measured in illumination (**a**, **c**, **e**) and dark (**b**, **d**, **f**) at different ambients: **a, b** in oxygen; **c, d** in nitrogen; **e, f** in vacuum

the annealing process. For example, the crystalline texture could be deteriorated when increasing annealing temperature [9], resulting in a lower relative intensity of $(\bar{4}02)$ diffraction peak. In addition, the relative intensities of diffraction peaks are almost unchanged for different annealing ambients if the annealing temperature is fixed. In other words, the annealing ambient has no significant influence on the crystalline texture of β-Ga_2O_3 thin film.

As shown in Fig. 2, the *I–V* characteristics of all the devices were measured in illumination and dark, respectively. It is found that the photoresponsivities of photoconductors decrease with the increase in annealing temperature. For example, both the photocurrents and the dark currents of photoconductors decrease significantly with the increase in annealing temperature at a voltage bias of 20 V. In particular, the photocurrents are reduced to about 1.2×10^{-12} A, close to the dark current, when the annealing temperature rises up to 1100 °C, and thus, the photoresponsivities of photoconductors are close to zero. It indicates that a higher annealing temperature can lead to a larger resistivity of β-Ga_2O_3 thin film. As discussed previously, the relative intensity of $(\bar{4}02)$ diffraction peak will decrease while increasing the annealing temperature, which means that the β-Ga_2O_3 thin film with a stronger relative intensity of $(\bar{4}02)$ diffraction peak should have lower resistivity and better photoresponsivity. The degradation of the photoresponsivity of photoconductor with the increase in annealing temperature is possibly due to the recrystallization effect as well, which makes the deterioration of the crystalline texture of β-Ga_2O_3 thin film.

Furthermore, Fig. 3 shows the *I–V* characteristics of the photoconductors based on the β-Ga_2O_3 thin films annealed at 800 °C. It is found that the photoconductor without any annealing exhibits the highest photocurrent and dark current compared with all the annealed samples. In addition, the vacuum-annealed photoconductor performs higher photocurrent and dark current than the oxygen- and nitrogen-annealed ones, and the lowest values of both parameters are observed in the oxygen-annealed one. It indicates that either nitrogen or oxygen annealing can increase the resistivity of β-Ga_2O_3 thin film, thus degrading the photoresponsivity of photoconductor. It was reported that the oxygen vacancies in β-Ga_2O_3 thin film normally act as donors and accordingly can release electrons to the conduction band [7, 10]. As for the vacuum-annealed sample, the concentration of the oxygen vacancies in β-Ga_2O_3 thin film is almost unchanged. However, the annealing for β-Ga_2O_3 thin film in oxygen increases the oxygen concentration and accordingly reduces the oxygen vacancies, resulting in a high resistivity and a low photoresponsivity. As for the nitrogen-annealed sample, the doping of nitrogen atoms, acting as acceptors, can be caused during the annealing process [11]. As a result, the recombination of the electrons and holes which are from oxygen vacancies and nitrogen doping, respectively, is induced, reducing the carrier concentration of β-Ga_2O_3 thin film and accordingly the photoresponsivity of the photoconductor based on such a material. In addition, the slow relaxation of photoconductor, which means that the signal cannot respond quickly enough to the switching on or off of the light source, often

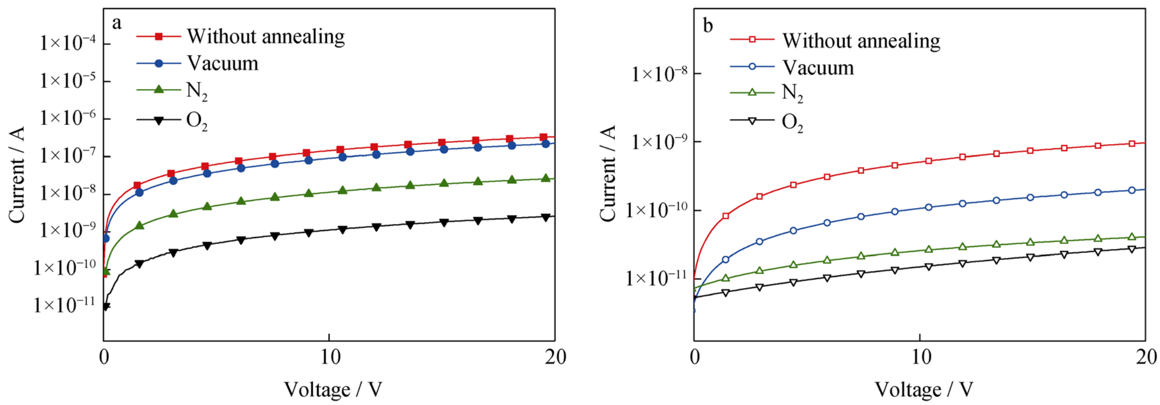

Fig. 3 *I–V* characteristics of β-Ga_2O_3 photoconductors before and after annealing at 800 °C measured in **a** illumination and **b** dark

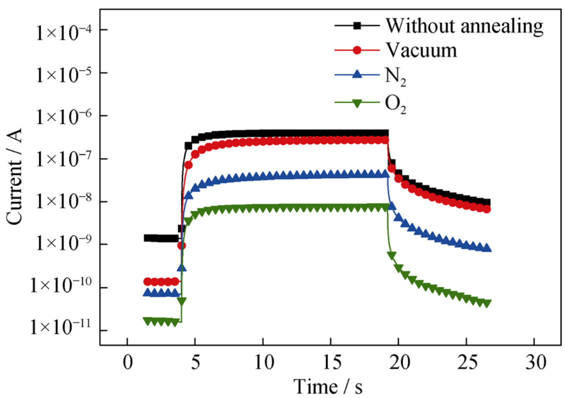

Fig. 4 Transient photoresponse of β-Ga_2O_3 photodetectors before and after annealing at 800 °C in different ambients

appears in its practical applications [12–14]. After removing the ultraviolet light source, the decay of the persistent photoconductivity (PPC) follows the exponential model expressed as follows:

$$i_{ph}(t) = i_{ph0}\exp(-t/\tau) \quad (1)$$

where $i_{ph}(t)$ is the instantaneous current at time (t), i_{ph0} is the current at the onset of decay, and it is the relaxation time. According to this equation, the time which elapses before $i_{ph}(t)$ decreasing to 1/e of i_{ph0} is called the falling time, and the time which elapses before $i_{ph}(t)$ rising to (e − 1)/e of i_{ph0} is called the rising time [15]. As shown in Fig. 4, the transient photoresponses of the photodetectors based on the β-Ga_2O_3 thin films before and after annealing at 800 °C in different ambients were measured. Meanwhile, both rising and falling time for each sample is listed in Table 1. It shows that the annealing for β-Ga_2O_3 thin film in oxygen can effectively improve the transient photoresponse and thus reduce the falling time of device. It could be attributed to the increase in the oxygen concentration in β-Ga_2O_3 thin film during oxygen annealing, which may lead to the decrease in the oxygen vacancies and point defects in β-Ga_2O_3 and reduce the PPC effect as a result.

Table 1 Rising and falling time of β-Ga_2O_3 photodetectors before and after annealing at 800 °C in different ambients

Treatment	Rising time/s	Falling time/ms
Without annealing	0.80	80
Annealing in vacuum	1.70	70
Annealing in nitrogen	1.90	80
Annealing in oxygen	0.85	20

4 Conclusion

In summary, the photoelectric properties of β-Ga_2O_3 thin film, which mainly depend on crystalline texture and oxygen vacancy, are influenced significantly by the annealing temperature and ambient in this work. It is found that the relative intensity of ($\bar{4}02$) diffraction peak is weakened with the increase in annealing temperature. Accordingly, the photoresponsivity of photoconductor decreases since the crystalline texture of β-Ga_2O_3 thin film is deteriorated. Moreover, the vacuum-annealed sample exhibits a higher photoresponsivity than the oxygen- and nitrogen-annealed samples even at the same annealing temperature, which is possibly attributed to a higher concentration of oxygen vacancies in β-Ga_2O_3 thin film. Furthermore, the oxygen annealing reduces the PPC effect of photoconductor significantly though the photoresponsivity is sacrificed.

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