High performance gate tunable solar blind ultraviolet phototransistors based on amorphous Ga$_2$O$_3$ films grown by mist chemical vapor deposition

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Abstract Gallium Oxide (Ga$_2$O$_3$) for solar-blind photodetectors (PDs) has drawing increasing research interest in recent years because of its natural wide bandgap. However, the traditional material growth methods are always complicated and the corresponding PD performance is also not good enough. In this work, amorphous Ga$_2$O$_3$ (a-Ga$_2$O$_3$) thin film grown by mist chemical deposition applied as solar blind deep ultraviolet phototransistors (PTs) is investigated for the first time, to solve the problem of high cost and time-consuming by traditional methods. Bottom-gate a-Ga$_2$O$_3$ three terminal thin film transistors (TFTs) are fabricated to boost their ultraviolet (UV) photodetection properties. Under the 254 nm UV illumination, the a-Ga$_2$O$_3$ PTs demonstrates a very high responsivity of 2300 $\text{AW}^{-1}$, external quantum efficiency of $1.12 \times 10^6 \%$ and detectivity of $1.87 \times 10^{14} \text{Jones}$. Such field-effect PTs with the ultrahigh performance address a significant step toward the feasibility and practicability of Ga$_2$O$_3$ PDs in the future applications.

KEYWORDS Ga$_2$O$_3$ thin film, mist-CVD, solar-blind phototransistor

1 | INTRODUCTION

In recent years, various key technology researches have been carried out around optoelectronic systems to realize the photodetectors (PDs) with high performance, low power consumption and low cost. Solar blind PDs (cut-off wavelength <280 nm) are one of the most promising applications. Due to the absorption of ozone in the atmosphere, ultraviolet (UV) light in the wavelength range of 200-280 nm is almost non-existent on the surface of the earth. Compared with other detection technologies, solar blind UV detection has the advantages of low false alarm rate, high sensitivity and high concealment. To date, the materials with bandgap tuned by alloy engineering such as AlGaN,$^{[1]}$ ZnMgO,$^{[2]}$ and ZnGaO$^{[3]}$ have been applied in PDs to realize the solar blind UV detection. However, the complex alloying process usually degrades the material crystallization quality (such as the composition fluctuations and phase separation) and thus limits the detection performance of the solar blind UV PDs.
Gallium oxide ($\text{Ga}_2\text{O}_3$) with a wide bandgap of about 4.7–5.2 eV, high radiation hardness, high thermal, chemical and physical stability,[4–10] is emerging as one of the promising candidate wide bandgap semiconductor materials. The optical absorption edge naturally corresponds to the wavelength at the deep UV spectral region (200-280 nm) without tuning its bandgap through complex and uncontrollable alloying process. Thus, it has attracted a lot of research attentions. Based on various kinds of $\text{Ga}_2\text{O}_3$ materials, different types of PDs have been demonstrated, which include metal-semiconductor-metal structures,[11] Schottky barrier diodes,[12] diamond/$\text{Ga}_2\text{O}_3$ heterojunctions,[13] and graphene/$\text{Ga}_2\text{O}_3$ structures.[14]

Although there have been some reports about solar-blind $\text{Ga}_2\text{O}_3$ PDs, the key factors of photodetection, such as responsivity and external quantum efficiency (EQE), are too low to realize practical applications. Moreover, the general two terminal devices do not provide excellent bias control over the channel current and photosensitivity. Compared to the two-terminal devices, phototransistors (PTs) are also important PDs due to their abilities of controlling the channel current by gate bias. Three-terminal PTs possess the intrinsic gain of transistors which can effectively control the channel current through gate voltage and leads to a huge photoconductive gain. However, very few $\text{Ga}_2\text{O}_3$ solar-blind PTs have been reported, and their performance still needs to be further improved. Moreover, to meet the requirement for practical applications, low-cost, stable and reliable experimental methods are also very significant for the application of $\text{Ga}_2\text{O}_3$ PTs.

There are some methods to grow $\text{Ga}_2\text{O}_3$ films, for example, molecular beam epitaxy (MBE),[15] metalorganic chemical vapor deposition (MOCVD),[16] halide vapor phase epitaxy (HVPE)[17] and aqueous-solution spin-coating[18] method. These techniques are generally time-consuming and expensive and the growth rate is relatively low. The spin coating method is simple and easy to implement, but it is more difficult to realize the uniformity of the film and the reliability of the quality process. Compared to above methods, non-vacuum mist chemical-vapor-deposition (mist-CVD) method is a very simple and cost-effective process with easy operation for growth process. As shown in Figure 1A, the mist-CVD system is usually composed of two parts, the source solution atomized droplet generation part and the heating zone film reaction growth part. Source solution in a tank is atomized to micrometer-size droplet by an ultrasonic generator. Then, mist droplets are transferred from the tank into the furnace with a carrier gas and the films are grown by the thermal decomposition. Gallium acetylacetonate can be used as the Ga source, and $\text{H}_2\text{O}$ is the primary oxygen source for the $\text{Ga}_2\text{O}_3$ growth. The gas of $\text{N}_2$ or $\text{O}_2$ is used as the carrier gas.
gas to transport the atomized droplets while ensuring the pure growth atmosphere of the film. In the mist deposition, the mist particles will be thermally evaporated in the gas atmosphere and the Gallium acetylacetonate will react with oxygen source at the substrate surface. Pyrolytic reaction on the substrate surface leads to the formation of films. This is the same as the CVD processes and the sources will migrate on the surface followed by the well-controlled formation of the film. Low equipment cost, fast growth rate and easy to control doping make the mist-CVD method a hot spot in recent years.[19–21] However, there is still no reports about Ga$_2$O$_3$ PTs based on Si substrates by using the mist-CVD method.

In this work, we demonstrate a-Ga$_2$O$_3$ films grown by the mist-CVD method on the cost-effective Si/SiO$_2$ substrates to realize the PTs with ultrahigh performance and gate tunable photodetection for the first time. By applying the appropriate back-gate voltage, the photoelectric performance and dark current of PTs can be controlled effectively. The responsivity of the a-Ga$_2$O$_3$ PTs can reach 2300 AW$^{-1}$, which corresponds to a very high quantum efficiency of $1.12 \times 10^{6}$%, a large detectivity of $1.87 \times 10^{14}$ Jones under a weak 254 nm light illumination of 70 $\mu$Wcm$^{-2}$. Excellent optoelectronic performance, simple and inexpensive production cost make the solar-blind a-Ga$_2$O$_3$ thin film field-effect PTs promising for the next-generation optoelectronic applications.

2 | RESULTS AND DISCUSSION

To optimize the growth condition, control experiments were conducted by adjusting the growth temperature $T_g$ (300-700°C). The grown Ga$_2$O$_3$ films herein are referred as Film-300, Film-400, Film-500, Film-600, Film-700 and the corresponding PTs are named as PT-300, PT-400, PT-500, PT-600 and PT-700 for the different $T_g$s from 300°C to 700°C. The schematic cross section and top views of a typical as-fabricated device are shown in Figure 1B.

The transmittance spectra with the wavelength from 200 to 800 nm for Ga$_2$O$_3$ films grown on quartz substrates at 300-700°C are shown in Figure 2A. All the films exhibit a clear absorption edge at the deep UV wavelength region around 250 nm and the transmittance of the Ga$_2$O$_3$ films in visible region reaches 80%. Figure 2B is the $(\alpha h \nu)^2$ as a function of photon energy. The absorption coefficient increases rapidly at the photon energy range around 4.7-5.26 eV. The absorption edge is shifted towards long wavelength region, indicating a reduced bandgap with the increasing growth temperature. It is found that the transmittance of Film-300 is not completely absorbed at the wavelength of 200 nm, which may be due to the thin film grown at a low temperature. Figure 2C exhibits the XRD curve of Film-400. There are no characteristic peaks other than substrate peaks, indicating the amorphous nature of the film. The same results also occur in the other Ga$_2$O$_3$
Thus, all the Ga$_2$O$_3$ thin films grown on the Si/SiO$_2$ substrates are amorphous (a-Ga$_2$O$_3$). The cross-section SEM image is shown in Figure 2D. The thickness of amorphous a-Ga$_2$O$_3$ Film-400 is about 268 nm, corresponding a growth rate over 10 nm min$^{-1}$.

The variation of a-Ga$_2$O$_3$ surface morphologies with different $T_g$ is revealed by atomic force microscopy (AFM) images in Figure 3. It can be found that the surface of a-Ga$_2$O$_3$ film is flat and uniform with an obvious nanoparticulate morphology at a low growth temperature. With the increasing growth temperature, the root-mean-square (RMS) roughness in a scanned area of $5 \times 5$ µm$^2$ gradually increases from 0.46 nm (300°C) to 10.83 nm (700°C), indicating that the surface morphology strongly depends on the growth temperature. Excessive growth temperature makes the atomic motion violent, and the surface roughness increases. The minimum RMS value of Film-400 is 0.385 nm, suggesting that the surface of the a-Ga$_2$O$_3$ film deposited on Si/SiO$_2$ substrate is relatively smooth. In brief, the deposited a-Ga$_2$O$_3$ films grown at low temperatures have more flat surfaces and high uniformity. The change of the surface roughness of the a-Ga$_2$O$_3$ films, which may affect the carrier transport, may be a factor of influencing the devices performance discussed below.

Figure 4A presents the X-ray photoelectric spectroscopy (XPS) spectra of the O1s core-level for Film-400. The O$^{2-}$ ions surrounded by metal atoms Ga is related to the peak at the binding energy of 530.3 ± 0.2 eV (O1). The highest binding energy peak (OIII) (531.6 eV) is related to the dissociated oxygen or OH species on the surface of the a-Ga$_2$O$_3$ films.[22] The medium binding energy peak (OII) (531.2 ± 0.2 eV) is associated with O$^{2-}$ ions in the oxygen-deficient regions. Thus, the area ratio of the oxygen-deficiency-related peak (OII) to the whole O1s peak (O1 + OII + OIII) was calculated to be OII/(O1 + OII + OIII) = 41% for Film-400, which reveals that there are many oxygen vacancies existing in the a-Ga$_2$O$_3$ film. The other intensity ratios for Film-500, Film-600 and Film-700 (see Figure S2) decrease from 39% to 35% when $T_g$ is increased from 400°C to 700°C. This result indicates that oxygen vacancies in the a-Ga$_2$O$_3$ film decreased with the increasing $T_g$. As shown in Figure 4B, with the increase of $T_g$, the binding energy of O1s moves towards higher energy level, indicating that oxygen elements in the film increase and oxygen defects decrease.[23] The Ga3d peak also shifts to the higher energy as $T_g$ increases in Figure 4C, due to the increase of Ga–O binding structure. It is worth noting that the characteristic peaks of O1s and Ga3d for Film-300 are weak and irregular, indicating that a too low growth temperature is not conducive to the growth of Ga$_2$O$_3$ film, which will directly affect the electrical performance of the device.

Based on above material characterizations, the mist-CVD deposited a-Ga$_2$O$_3$ films were then used to fabricate bottom-gate, top-contact thin film transistors. From the dark I-V data shown in Figure S3, PT-300 and PT-700 exhibit poor electrical properties (on/off ratio only 2 orders) and the corresponding saturation drain current $I_{DS}$ are only 5 and 15 nA at a gate voltage ($V_{GS}$) 40 V and a drain biasvoltage ($V_{DS}$) of 10 V. A too low growth temperature can result in the insufficient energy for the Ga-O binding
bond, which is not conductive to the formation of a Ga$_2$O$_3$ film. This is consistent with the XPS analysis above. As for the PT-700, the low $I_{DS}$ may be due to the too rough surface of the film (RMS 10.83 nm) which will decrease the carrier migration. Figure 5A displays the transfer characteristics of PT-400, PT-500, and PT-600 for $V_{GS}$ from $-30$ to 40 V and a constant $V_{DS}$ of 10 V. All curves show that all the devices possess the obvious gate control capability. The on-state $I_{DS}$ increases rapidly as $V_{DS}$ increases. With an applied gate voltage $V_{GS}$ of 40 V and $V_{DS} = 10$ V, it is found that the saturation drain current $I_{DSat}$ are 12 $\mu$A for PT-400 and 5 $\mu$A for PT-500. In contrast, $I_{DS}$ is only 40 nA for PT-600 under the same bias condition. The corresponding transistor electrical parameters (such as the threshold voltage $V_{TH}$, ON/OFF ratio, subthreshold swing SS, and electron mobility $\mu$) can be determined from the dark I–V data. $V_{TH}$ and $\mu$ can be determined from the linear plot of $I_{DS1/2}$ versus $V_{GS}$ according to the following equation:

$$I_{DS} = \left( \frac{\mu W C_{ox}}{2L} \right) (V_{GS} - V_{TH})^2$$

(1)

where $W$ is the channel width, $L$ is the channel length, and $C_{ox}$ is the capacitance per unit area of the gate dielectric. SS can be extracted from the linear region of the transfer curve by

$$SS = \frac{dV_{GS}}{d(logI_{DS})}$$

(2)

Table 1 lists the electrical parameters extracted for PT-400, PT-500, and PT-600. The extracted mobility ($\mu$) of the optimal PT-400 is only 0.1 cm$^2$ V$^{-1}$s$^{-1}$, which may be due to the amorphous nature of Ga$_2$O$_3$ material. In spite of this, the device still exhibits excellent electrical performance, such as on/off ratio of $\approx 10^4$, a subthreshold swing of 0.125 V dec$^{-1}$ and a threshold voltage ($V_{th}$) of $-5$ V. Thus, the photoelectric properties of this device are carefully investigated in the following.

Xenon lamp light source was used to produce 254 nm UV light irradiate vertically on the a-Ga$_2$O$_3$ PTs to study the photoelectric performance at room temperature. The incident light power is calibrated before the measurements by a standard silicon PD. Figure 5B shows the transfer characteristics of the PT-400 measured under 254 nm illumination with different light intensities. The incident light power is calibrated before the measurements by a standard silicon PD. Figure 5B shows the transfer characteristics of the PT-400 measured under 254 nm illumination with different light intensities. The incident light power is calibrated before the measurements by a standard silicon PD. Figure 5B shows the transfer characteristics of the PT-400 measured under 254 nm illumination with different light intensities. The incident light power is calibrated before the measurements by a standard silicon PD. Figure 5B shows the transfer characteristics of the PT-400 measured under 254 nm illumination with different light intensities.
FIGURE 5  A, Transfer characteristics of PT-400, PT-500, and PT-600 measured in dark with a constant VDS of 10 V. B, Transfer characteristics of PT-400 under 254 nm light illumination with different light intensities. (C) Transfer characteristics of PT-400 for different VDS under dark and 254 nm light illumination with the intensity of 70 µWcm$^{-2}$. D, IDS-VDS output characteristics for different VGS in dark and 254 nm light illumination with the intensity of 70 µWcm$^{-2}$. E, Time-dependent photoresponse characteristics of the a-Ga$_2$O$_3$ PT with 15 dark-light cycles. F, Time-dependent photoresponse characteristics with various VDS. G, Time-dependent photoresponse characteristics at different illumination intensities.
attributed to the carrier generation from the band-to-band transition in the a-Ga$_2$O$_3$ layer. It should be noted that a large negative shift in turn-on voltage is observed with UV illumination, which is attributed to the large number of photo-generated carriers in the channel. The photogenerated carriers induce a large photocurrent, making the applied gate bias lose its control on the transfer operation. Figure 5C shows the transfer characteristics of PT-400 measured under 254 nm illumination with various $V_{DS}$ (10, 20, and 30 V). The change of photocurrent compared to dark current (about 1 nA) becomes more and more obvious with the increase of $V_{DS}$. Figure 5D shows the $I_{DS}$-$V_{DS}$ curves of the PT-400 under dark and 254 nm UV illumination conditions. The photocurrent increases as $V_{GS}$ increases from $-5$ to 40 V because of higher gate bias will generate much stronger electric field, which will produce more charges in the channel. Figure 5E shows a-Ga$_2$O$_3$ PT-400 time-dependent photoelectric response characteristics. It shows that the device can be well switched between on and off states after 15 dark-light cycles, meaning a good device repeatability. The rise time ($\tau_r$) and decay time ($\tau_d$) are estimated to 10 and 6 seconds, respectively. The long response time might result from the persistent photoconductivity effect caused by large number of oxygen vacancies and high density of trap states. Defects act as carrier capture centers, capturing and releasing carriers; thus, increasing device response time. Figure 5F shows the time-dependent photoelectric response characteristics of the PT-400 at $V_{GS} = 10$ V and different $V_{DS}$ in the range from 5 to 15 V. The PT possesses high stability and fine repeatability because the device exhibits almost the identical response after multiple illumination cycles under the 254 nm UV light intensity of 70 $\mu$W/cm$^2$. It can be clearly seen that the photocurrent increases with the increasing $V_{DS}$, and this is because a larger applied voltage will lead to a higher electric field, and a higher electric field can accelerate the velocity of carriers and then improve the collection of the photo-generated carriers by the electrodes, and thus the photocurrent is significantly enhanced. The time-dependent photoresponse under different illumination powers under 20 V bias is shown in Figure 5G.

The photocurrent increases with the increasing light intensity because more photons can be absorbed and more photoexcited carriers can be generated in the a-Ga$_2$O$_3$ film under a higher light intensity. It also shows that the device can be well switched between on and off states with good reproducibility. The mechanism of the phototransistors can be further interpreted by the schematic and the energy band diagrams of the a-Ga$_2$O$_3$ thin film phototransistor under dark and 254 nm illumination at different $V_{GS}$ shown in Figure 6A-D. The device was fully depleted with a low off current since neither electric channel nor photo-generated carriers form when $V_{GS} < V_{th}$ in the dark. When the device is illuminated with UV light, a large number of electron–hole pairs could be generated in the channel through intrinsic valence band ($E_v$) to conduction band ($E_C$) transition. The deep-level neutral oxygen vacancy $V_o$ is ionized to shallow donors $V_o^{2+}$ or $V_o^+$ both of which make a big contribution to the current flowing between source and drain electrodes. The photoexcited carriers will be separated and then drift between drain and source contacts under a moderate $V_{DS}$. In addition, the carriers accumulated at the a-Ga$_2$O$_3$–SiO$_2$ interface get energy from the applied drain to gate electric field and surpass the barrier height to move into the channel. Therefore, carriers are injected into the channel by surpassing the barrier height due to the applied electric field. The large number of carriers injected into the channel collecting as the source-drain to generate a large photocurrent. After the UV light is turned off, the photogenerated electrons caused by the gate bias recover back to the neutral $V_o$ states, making the device still maintain the low current.

Responsivity (R), Detectivity ($D^*$) and External Quantum Efficiency (EQE) are the key parameters to evaluate the performance of the PT. R is defined as

$$R = (I_{photo} - I_{dark})/(P_\lambda S)$$

where $S$ is the effective illumination area of the device. R can reach as high as 2300 $AW^{-1}$ at $V_{GS} = -10$ V and $V_{DS} = 10$ V under a UV light 70 $\mu$W cm$^{-2}$ illumination. The

| Materials     | Method      | R [AW$^{-1}$] | EQE(%) | D(Jones) | Ref     |
|---------------|-------------|--------------|--------|----------|---------|
| a-Ga$_2$O$_3$ | Mist-CVD    | 2300         | 1.12 $\times 10^6$ | 1.87 $\times 10^{14}$ | This work |
| a-Ga$_2$O$_3$ | Sputter     | 70.26        | ~      | 1.26 $\times 10^{14}$ | [24]     |
| a-Ga$_2$O$_3$ | ALD         | 45.1         | ~      | ~        | [25]     |
| $\beta$-Ga$_2$O$_3$ | Sputter | 96           | 4.768 $\times 10^4$ | ~        | [26]     |
| $\beta$-Ga$_2$O$_3$ | MBE    | 54.9         | ~      | 3.71 $\times 10^{14}$ | [27]     |
| a-Ga$_2$O$_3$ | Sputter     | 4100         | 2 $\times 10^5$  | 2.5 $\times 10^{13}$ | [28]     |
| $\alpha$-Ga$_2$O$_3$ | MOCVD | 11.5         | ~      | 1 $\times 10^{15}$ | [29]     |
All these show that the fabricated PT has the excellent photoelectric properties at different light intensities. The excellent optoelectronic performance, simple and inexpensive production cost make the solar-blind a-Ga$_2$O$_3$ thin film field-effect PTs promising for the next-generation optoelectronic applications.

3 | CONCLUSION

In this paper, we have grown a-Ga$_2$O$_3$ thin film by using the low-cost mist-CVD method under different temperatures on Si/SiO$_2$ substrates. The deposited a-Ga$_2$O$_3$ films possess a good quality with a flat surface and high uniformity at the low growth temperature of 400°C and the RMS value is as low as 0.385 nm. The PT based on the optimal a-Ga$_2$O$_3$ film is highly sensitive to 254 nm UV light with good reproducibility and stability under different bias voltages and light intensities. An ultrahigh $R$ of 2300 A W$^{-1}$, detectivity D$^*$ of $1.87 \times 10^{14}$ Jones, EQE of $1.12 \times 10^6\%$ were achieved at $V_{GS} = -10$ V, $V_{DS} = 10$ V under the UV light intensity of 70 $\mu$W cm$^{-2}$. The simple and inexpensive growth method and the excellent optoelectronic performance of the fabricated PTs demonstrate that the mist-CVD assisted Ga$_2$O$_3$ thin film may be implemented as a good semiconductor material for the solar-blind PDs for the next-generation optoelectronic applications.
EXPERIMENTAL SECTION

Gallium acetylacetonate (Ga(Acac)₃), 99.99%, was acquired from Alfa. Hydrochloric acid, 30%, was purchased from Wako Pure Chemical Industries. A 0.05 M solution was prepared from 100 mL distilled H₂O, 1 mL HCl and 1.85 g Ga(Acac)₃. The solution was atomized via ultrasonication at a frequency of 1.7 MHz. The mist particles were carried by N₂ gas with a rate of 3 L min⁻¹ to a commercial (100) p-type Si/SiO₂ wafers (MK NANO CO., LTD) with a 200 nm SiO₂ layer as the gate dielectric, on which the a-Ga₂O₃ film was grown for 20 minutes at different temperatures (300-700°C) in a furnace. The highly boron doped Si wafers with a resistivity between 0.01 and 0.05 Ω cm acted as the common bottom gate electrode. Ti/Au (20/100 nm) metal was deposited using electron beam evaporation for contacts. The metal electrodes had an interdigitated geometry with 12 fingers: 80 µm long, 5 µm wide, and 5 µm finger spacing with an active area of 5350 µm².

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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