(In$_x$Ga$_{1-x}$)$_2$O$_3$ Photodetectors Fabricated on Sapphire at Different Temperatures by PLD

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Abstract: The (In$_x$Ga$_{1-x}$)$_2$O$_3$ photodetectors were fabricated on the single-crystalline (In$_x$Ga$_{1-x}$)$_2$O$_3$ films deposited on sapphire substrate by pulsed laser deposition. The structural and optical properties of the epilayers were investigated using high-resolution X-ray diffraction, X-ray photoelectron spectroscopy, spectroscopic ellipsometry, and transmittance spectra. With decreasing the growth temperature, the indium composition increased and the bandgap decreased from 4.99 eV to 4.89 eV (In$_{0.05}$Ga$_{0.95}$)$_2$O$_3$ and 4.78 eV (In$_{0.08}$Ga$_{0.92}$)$_2$O$_3$. Furthermore, the photoelectrical characteristics of (In$_x$Ga$_{1-x}$)$_2$O$_3$ detectors were also studied. The enhanced $I_{photo}$, $I_{dark}$, and responsivity $R$ were achieved in the devices with higher In composition, while a larger number of defects were introduced, resulting in the significant persistent photoconductivity.

Index Terms: (In$_x$Ga$_{1-x}$)$_2$O$_3$, photodetectors, temperatures, PLD.

1. Introduction

Recently, Ga$_2$O$_3$ has attracted great attention for high power electronic and ultraviolet (UV) or deep-ultraviolet (DUV) applications because of its exceptional properties, such as large bandgap ($E_G$), high theoretical breakdown electric field and physical and chemical stabilities, etc. [1]–[5]. There exist five polymorphs of Ga$_2$O$_3$ ($\alpha$, $\beta$, $\gamma$, $\delta$ and $\epsilon$ phases), among which $\beta$-Ga$_2$O$_3$ is the most common and most stable crystal. $\beta$-Ga$_2$O$_3$ photodetectors have been fabricated and characterized with the cutoff wavelength around 250 nm [6]–[8]. It is known that the $E_G$ can be tuned by incorporating Al or In into $\beta$-Ga$_2$O$_3$ to broaden the photodetection range. (In$_x$Ga$_{1-x}$)$_2$O$_3$ films have been deposited by various methods: metal organic chemical vapor deposition (MOCVD) [9], molecular beam epitaxy (MBE) [10], Sol-gel [11], and pulsed laser deposition (PLD) [12]. So far, a few studies have been carried out on the growth of (In$_x$Ga$_{1-x}$)$_2$O$_3$ epilayers and the fabrication of (In$_x$Ga$_{1-x}$)$_2$O$_3$ photodetectors [11], [13].

In this paper, the single-crystalline (In$_x$Ga$_{1-x}$)$_2$O$_3$ films were deposited on c-plane sapphire substrates by PLD. The properties of the (In$_x$Ga$_{1-x}$)$_2$O$_3$ epilayers were investigated by High resolution X-ray diffraction (HRXRD), X-ray photoelectron spectroscopy (XPS), spectroscopic ellipsometry
(In\textsubscript{x}Ga\textsubscript{1-x})\textsubscript{2}O\textsubscript{3} photodetectors were fabricated and characterized with different In composition. Compared with Ga\textsubscript{2}O\textsubscript{3} photodetectors, the cutoff wavelength of (In\textsubscript{x}Ga\textsubscript{1-x})\textsubscript{2}O\textsubscript{3} devices could be tuned from 253 nm (x = 0) to 335 nm (x = 1), which may be widely used in military and civil applications, such as flame detection, early missile threat warning. In our work, the better performance has been achieved in (In\textsubscript{x}Ga\textsubscript{1-x})\textsubscript{2}O\textsubscript{3} photodetectors, including photocurrent and responsivity, although the cutoff wavelength only changes from 249 nm (Ga\textsubscript{2}O\textsubscript{3} on sapphire substrate) to 254 nm (x = 0.05) and 260 nm (x = 0.08).

2. Experiments

(In\textsubscript{x}Ga\textsubscript{1-x})\textsubscript{2}O\textsubscript{3} films were grown on double-polished (0001)-oriented sapphire substrates using PLD method. The ceramic (In\textsubscript{x}Ga\textsubscript{1-x})\textsubscript{2}O\textsubscript{3} with In composition of x = 0.10 was used as target. The KrF excimer laser with a frequency of 3 Hz and the energy density of 2.0 J/cm\textsuperscript{2} was applied to irradiate the target. The (In\textsubscript{x}Ga\textsubscript{1-x})\textsubscript{2}O\textsubscript{3} epilayers were deposited in a lower oxygen pressure of 0.003 mbar at the temperatures of 500 °C (sample A) and 550 °C (sample B), respectively. The Ga\textsubscript{2}O\textsubscript{3} epilayer was also grown as a reference (Sample C). For the photodetectors fabrication, the interdigital Ti/Au (10 nm/100 nm) films were deposited using electron beam evaporation as the schottky contacts, with the finger spacing of 100 μm and the total length of 17.5 mm.

3. Results and Discussion

3.1 Material Characterization

The structural properties of the samples were investigated by HRXRD using CuK\textsubscript{α} radiation at room temperature. Fig. 1 shows the HRXRD curves of the three samples. There are four diffraction peaks of (In\textsubscript{x}Ga\textsubscript{1-x})\textsubscript{2}O\textsubscript{3} and Ga\textsubscript{2}O\textsubscript{3} samples on sapphire. The peaks of (In\textsubscript{x}Ga\textsubscript{1-x})\textsubscript{2}O\textsubscript{3} are located at the left side of Ga\textsubscript{2}O\textsubscript{3} peaks.
Fig. 2. XPS spectra of (a) In3d5/2, (b) O1s and (c) Ga2p3/2 for (In$_x$Ga$_{1-x}$)$_2$O$_3$ and Ga$_2$O$_3$ samples.

The XPS peaks corresponding to the $(\bar{2}01)$, $(402)$ and $(603)$ planes of $\beta$-(In$_x$Ga$_{1-x}$)$_2$O$_3$ and sapphire substrate (0006) planes, respectively. The (In$_x$Ga$_{1-x}$)$_2$O$_3$ peaks are located at the lower angle side in comparison with the Ga$_2$O$_3$ peaks, demonstrating that In atoms have been incorporated into the lattice of $\beta$-Ga$_2$O$_3$.0. In addition, the peaks of (In$_x$Ga$_{1-x}$)$_2$O$_3$ shift to lower angle as the temperature decreasing to 500 °C, indicating more In atoms in sample A than sample B. According to Bragg’s equation, the interplanar spacings $d$ of $(\bar{2}01)$, $(402)$ and $(603)$ planes for the three samples are 0.4726, 0.2368 and 0.1578 nm (sample A), 0.4706, 0.2354 and 0.1570 nm (sample B), 0.4594, 0.2348 and 0.1566 nm (sample C), in good agreement with (In$_x$Ga$_{1-x}$)$_2$O$_3$ films deposited by continuous composition spread (CCS) PLD [15].

Fig. 2 presents the XPS peaks of In3d5/2, O1s and Ga2p3/2 for (In$_x$Ga$_{1-x}$)$_2$O$_3$ and Ga$_2$O$_3$ samples. Before XPS measurement, the surface of the samples was etched by in situ Ar$^+$ ion and the binding energy was calibrated using the C1s line (284.6 eV) from adventitious carbon. With In composition increment, In3d5/2 peak shifts toward In3d5/2 of In$_2$O$_3$ (444.5 eV) and Ga2p3/2 peak deviates from Ga2p3/2 of Ga$_2$O$_3$ (1118.3 eV), as shown in Fig. 2(a) and (c), indicating that Ga atoms have been effectively substituted by In atoms [16]. Based on the area of the peaks and the corresponding sensitivity factors, In composition of (In$_x$Ga$_{1-x}$)$_2$O$_3$ films are calculated to be 8% and 5% for the sample A and B. Fig. 2(b) shows the Gaussian fitting results of O1s peaks. The oxygen vacancies can be determined by calculating the area ratio (O$_{\text{II}}$/O$_{\text{I}}$ + O$_{\text{II}}$), where O$_{\text{I}}$ and O$_{\text{II}}$ are associated with O$^{2-}$ ions surrounded by Ga/In atoms and oxygen vacancies in the epilayers [13], [17]. The ratios are about 13% and 6% for sample A and B, indicating more oxygen vacancies introduced in the sample A.

The variable-angle spectral ellipsometry was used to determine the thicknesses, refractive index $n$ (Fig. 4(a)) and extinction coefficient $k$ (Fig. 4(b)) at room temperature. The incidence angles were set to 55°, 65° and 75° in the 200–800 nm spectral range. The Cauchy dispersion fitting and point-by-point method were applied for 400–800 nm and 200–400 nm spectra range, respectively [18], [19]. Fig. 3 presents the fitting results with the thickness of 358, 320 and 198 nm for the three samples. The refractive index $n$ increases with the In composition, which is similar to the reference [19]. The extinction coefficient $k$ of (In$_x$Ga$_{1-x}$)$_2$O$_3$ epilayers is higher than that of Ga$_2$O$_3$ control and the absorption edge shifts toward longer wavelength as Fig. 4(b) shown, indicating that (In$_x$Ga$_{1-x}$)$_2$O$_3$ films have narrower bandgap depending on the In composition $x$. For the higher extinction coefficient $k$ of (In$_x$Ga$_{1-x}$)$_2$O$_3$ sample, the thickness can be reduced in comparison with
Fig. 3. SE measured data of $\Psi$ (left) and $\Delta$ (right) and the fitting line of $(\text{In}_{x}\text{Ga}_{1-x})_2\text{O}_3$ and $\text{Ga}_2\text{O}_3$ samples at angles of 55°, 65° and 75°.

Fig. 4. Spectra of (a) the refractive index ($n$) and (b) the extinction coefficient ($k$) for the three samples and the inset is $E_G$ for different samples calculated by the extinction coefficient.
Ga$_2$O$_3$ film to adequately absorb the incident light. The Ga$_2$O$_3$ and (In$_x$Ga$_{1-x}$)$_2$O$_3$ photodetectors can be both widely used in the civil and military applications, such as missile tracking, fire detection, ozone holes monitoring, chemical/biological analysis. The $E_G$ of (In$_x$Ga$_{1-x}$)$_2$O$_3$ films can be obtained by extrapolating the linear region of $(\alpha h\nu)^2 h\nu$ to the horizontal axis, where $h\nu$ is the energy of the incident photon and $\alpha$ is the absorption coefficient of the films, which can be calculated by the relation $\alpha = 4\pi k/\lambda$. Therefore, the bandgap $E_G$ can be found to be 4.78, 4.89 and 4.99 eV, respectively, as depicted in the inset of Fig. 4(b).

The optical transmittance spectra of the samples are shown in Fig. 5. The average transmittance is beyond 70% in the visible region and a sharp absorption edge can be observed. With increasing In content, the absorption edge shifts toward longer wavelength. In addition, the absorption coefficient $\alpha$ can also be calculated by the relation $\alpha = (1/t)\ln[(1 - R_c)^2/T]$ [20], where $t$ is the thickness and $R_c$ and $T$ are the reflectance and transmittance, respectively. The thicknesses of the epilayers were determined by SE discussed above. For the transmittance greater than 70% with the $\lambda$ longer than 400 nm, the reflectance about 30% was adopted to determine $\alpha$. Then $E_G$ of the three samples are 4.80, 4.91, and 5.00 eV, respectively, in good agreement with the SE results.

### 3.2 Photoelectric Characteristics of the Devices

The photoelectric characteristics of (In$_x$Ga$_{1-x}$)$_2$O$_3$ photodetectors were investigated using a UV lamp, including the dark current $I_{\text{dark}}$, photocurrent $I_{\text{photo}}$, time-dependent photoresponse and the responsivity $R$. Fig. 6(a)–(c) represents the $I_{\text{photo}}$ versus the bias voltage $V_{\text{bias}}$ with a normalized optical power density $P_{\text{light}}$ of 0, 300, 600 and 900 $\mu$W/cm$^2$ and (d) is the dark current $I_{\text{dark}}$ plotted on a logarithmic scale. The $I_{\text{dark}}$ and $I_{\text{photo}}$ of (In$_x$Ga$_{1-x}$)$_2$O$_3$ devices are higher than that of Ga$_2$O$_3$. The larger $I_{\text{dark}}$ may be related to the enhancement of (In$_x$Ga$_{1-x}$)$_2$O$_3$ conductivity [21]. During illumination, a large number of electron-hole pairs would be generated and the holes drift to cathode electrode, resulting in the decrease of Schottky barrier height at the ground contact, more electrons injection into the channel. In addition, the effective Schottky barrier height of (In$_x$Ga$_{1-x}$)$_2$O$_3$ devices
Fig. 6. The linear photocurrent for (a) (In$_{0.08}$Ga$_{0.92}$)$_2$O$_3$, (b) (In$_{0.05}$Ga$_{0.95}$)$_2$O$_3$ and (c) Ga$_2$O$_3$ devices under 254 or 260 nm illumination with various $P_{\text{light}}$ and (d) The logarithmic dark current $I_{\text{dark}}$ versus $V_{\text{bias}}$ for the three samples.

Fig. 7. Time-dependent $I_{\text{photo}}$ characteristics and the fitting line of the rise and decay process for different photodectors at $V_{\text{bias}} = 20$ V and $\lambda = 254$ or 260 nm.

will also be reduced with In composition [22], [23]. As a result, more extra carriers can be collected, leading to the boosting $I_{\text{photo}}$.

Fig. 7 depicts the typical time-dependent photoresponse characteristics of fabricated photodetectors. During the measurements, an illumination $\lambda$ of 254 nm (or 260 nm) square-wave light was used with $V_{\text{bias}}$ of 10 V, $P_{\text{light}}$ of 300 $\mu$W/cm$^2$ and period of 40 s. The rise and decay processes...
Fig. 8. \( R \) versus illumination wavelengths \( \lambda \) for the different photodetectors at \( V_{\text{bias}} = 15 \) and 30 V.

consist of two procedures, fast-response and slow-response. It is known that the defects levels in the bandgap might trap the photogenerated carriers and delay the carriers collection during UV light illumination or the recombination after switching off the light, leading to the slow-response, while the fast-response is associated with the band to band transition 0. The time constants \( \tau_1 \) and \( \tau_2 \) were determined to be 0.63(0.65@260 nm), 0.67, 0.82 sec and 6.59(6.68@260 nm), 5.19, 5.03 sec and those of decay process \( \tau_d1 \) and \( \tau_d2 \) were 0.52(0.57@260 nm), 0.48, 0.39 sec and 8.69(8.75@260 nm), 8.04, 5.73 sec for (In\(_{0.08}\)Ga\(_{0.92}\))\(_2\)O\(_3\), (In\(_{0.05}\)Ga\(_{0.95}\))\(_2\)O\(_3\) and Ga\(_2\)O\(_3\) photodetectors, respectively. The larger time constant \( \tau_d2 \) of (In\(_{0.08}\)Ga\(_{0.92}\))\(_2\)O\(_3\) suggests there are a greater number of defects in the bandgap, resulting in a larger persistent photoconductivity (PPC).

The \( R \) as a function of the illumination wavelengths \( \lambda \) for the fabricated devices is shown in the Fig. 8. A higher \( R \) was obtained in (In\(_{x}\)Ga\(_{1-x}\))\(_2\)O\(_3\) photodetectors, compared with Ga\(_2\)O\(_3\) device. The maximum \( R \) (\( R_{\text{max}} \)) are 6.12A/W, 0.48A/W and 0.07A/W at the \( V_{\text{bias}} \) of 30 V for the three samples and the \( R_{\text{max}} \) shifts toward longer wavelengths with increasing In content. According to the equation \( E_G = hc/\lambda_c \), where \( h \) is Planck’s constant, \( c \) is velocity of light and \( \lambda_c \) is the cut-off wavelength corresponding to the direct bandgap 0, \( E_G \) of the fabricated photodetectors are calculated to be 4.77, 4.88 and 4.98 eV, respectively, illustrating the decrease of (In\(_{x}\)Ga\(_{1-x}\))\(_2\)O\(_3\) bandgap of for higher In composition, which is consistent with the transmittance and SE results.
4. Conclusion

We deposited single-crystal \((\text{In}_x\text{Ga}_{1-x})_2\text{O}_3\) films on c-plane sapphire substrates using PLD. Compared with \(\text{Ga}_2\text{O}_3\), the bandgap and conductivity is important for the \((\text{In}_x\text{Ga}_{1-x})_2\text{O}_3\) film used for photodetectors. With the In composition increasing, the bandgap decrease and the cutoff wavelength will be red-shifted or shift to the lower energy. On the other hand, the larger the In composition, the higher the dark current and the enhanced \(I_{\text{photo}}\) and \(R\), which is attributed to the incorporation of indium leading to the higher conductivity and lower Schottky barrier height. With the \((\text{In}_x\text{Ga}_{1-x})_2\text{O}_3\) growth temperature decreasing, a higher In composition was obtained and a larger number of defects were also introduced into the \((\text{In}_x\text{Ga}_{1-x})_2\text{O}_3\) films, resulting in the obvious PPC.

References


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