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Two-step growth of β-Ga_2O_3 on *c*-plane sapphire using MOCVD for solar-blind photodetector

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Abstract: In this work, a two-step metal organic chemical vapor deposition (MOCVD) method was applied for growing β-Ga_2O_3 film on *c*-plane sapphire. Optimized buffer layer growth temperature (T_B) was found at 700 °C and the β-Ga_2O_3 film with full width at half maximum (FWHM) of 0.66° was achieved. A metal–semiconductor–metal (MSM) solar-blind photodetector (PD) was fabricated based on the β-Ga_2O_3 film. Ultrahigh responsivity of 1422 A/W @ 254 nm and photo-to-dark current ratio (PDCR) of 10^6 at 10 V bias were obtained. The detectivity of 2.5×10^{15} Jones proved that the photodetector has outstanding performance in detecting weak signals. Moreover, the photodetector exhibited superior wavelength selectivity with rejection ratio ($R_{250\ nm}/R_{400\ nm}$) of 10^5. These results indicate that the two-step method is a promising approach for preparation of high-quality β-Ga_2O_3 films for high-performance solar-blind photodetectors.

Key words: MOCVD; two-step growth; β-Ga_2O_3; solar-blind photodetector; responsivity

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1. Introductions

As an emerging wide band gap semiconductor material, β-Ga_2O_3 is considered as one of the most promising materials for power-electronic devices and solar-blind detectors. It is benefit from the excellent material properties of β-Ga_2O_3 including ultrawide bandgap (4.6–5.2 eV), high critical field (8 MV/cm), high Baliga's figure-of merit (3214) and good chemical stability[1−3]. β-Ga_2O_3 solar-blind ultraviolet (200–280 nm) photodetectors have wide application owing to their superiorities, such as low background noise, high response value and radioresistance[4, 5]. In order to improve the photodetector performance, a series of methods are conducted to grow β-Ga_2O_3 films, including molecular beam epitaxy (MBE)[6], halide vapor phase epitaxy (HVPE)[7], low pressure chemical vapor deposition (LPCVD)[8, 9], mist chemical vapor deposition (mist CVD)[10] and MOCVD[11]. Metal–semiconductor–metal (MSM) structure has the advantages of simple fabrication process, convenient integration with readout circuits and high responsivity because of internal gain. Pratiyush *et al*.[12] demonstrated MSM detector using heteroepitaxial β-Ga_2O_3 films with a high responsivity of 1.5 A/W. Qian *et al*.[13] reported MSM detector based on amorphous Ga_2O_3 films with an ultrahigh response of 70.26 A/W and specific detection rate of 1.26×10^{14} Jones. By growing polycrystalline β-Ga_2O_3 film, Arora *et al*.[14] had prepared the high-performance MSM detector with a response of 96.13 A/W.

Although β-Ga_2O_3 film grown by various technologies have been reported, there are still some difficulties in getting ideal quality β-Ga_2O_3 films. For example, the expensive substrate for homoepitaxial and the large lattice mismatch for heteroepitaxial. As low-cost substrates, sapphires are widely used in heteroepitaxial β-Ga_2O_3 films. However, the lattice mismatch between *c*-sapphire and β-Ga_2O_3 is 3.5%. The stress caused by lattice mismatch will lead to a high dislocation density, which is not conducive to the epitaxial growth of material. In order to alleviate lattice mismatch and improve crystalline quality of epitaxial films, a buffer layer between substrate and epitaxial layer can be applied. However, there are few reports about the effect of buffer layer on the quality of β-Ga_2O_3 film on *c*-sapphire[15, 16].

In this work, high-quality β-Ga_2O_3 films were grown by two-step MOCVD method on *c*-sapphire. By varying buffer layer growth temperature, β-Ga_2O_3 film with fullwidth at half maximum (FWHM) of 0.66° was obtained. The MSM solar-blind photodetector was fabricated to further verify the excellent performance of β-Ga_2O_3 film quality. The solar-blind photodetector exhibits high responsivity of 1422 A/W @ 254 nm. In addition, ultrahigh PDCR (10^6), detectivity (2.5×10^{15} Jones) and respond speed (36 ms) were also achieved from the fabricated photodetector. This work provides an approach for the preparation of high-quality β-Ga_2O_3 films and the application of β-Ga_2O_3 solar-blind photodetectors.

2. Experimental details

Four β-Ga_2O_3 samples were prepared by depositing on *c*-plane (0001) sapphire in a MOCVD system by the two-step method. First, the thin buffer layer of Ga_2O_3 was deposited at low temperature (T_B), and then β-Ga_2O_3 film was grown on the buffer layer at 960 °C. Trimethylgallium (TMG) and O_2 were used as gallium and oxygen precursors, respectively. High-purity argon was used as carrier gas for TMG source. Four β-Ga_2O_3 films samples were grown at the same condition, except that T_B were at 650 , 700 , 750 and 800 °C. The crystalline quality of β-Ga_2O_3 films were characterized by high-reso-

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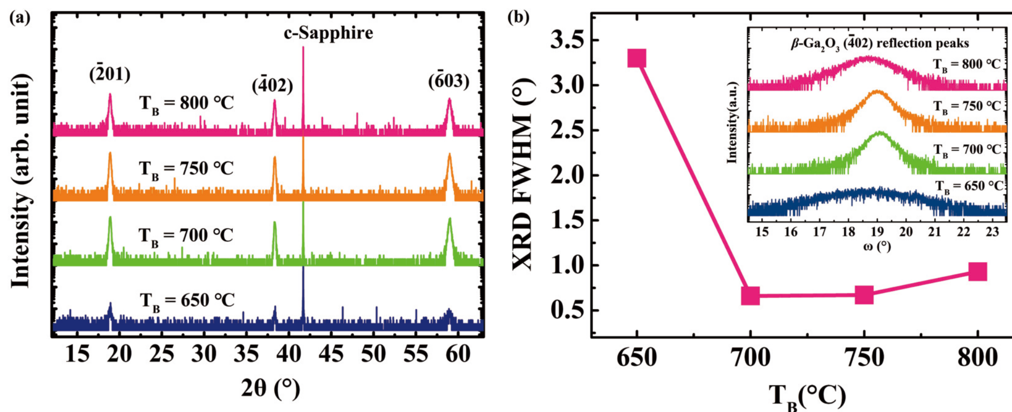

Fig. 1. (Color online) (a) XRD ω–2θ scan and (b) FWHM of rocking curve for β-Ga_2O_3 films grown at different T_B. Inset shows the XRD ω scan for films.

lution X-ray diffraction (HRXRD, X'Pert3 MRD). The surface morphologies of films were investigated by atomic force microscopy (AFM, SII Nanonavi) and scanning electron microscope (SEM, NanoSEM650). The photodetector with MSM structure was fabricated on the β-Ga_2O_3 film with T_B = 700 °C. Si ion implantation process[17, 18] was performed and followed by deposition of Ti/Au electrode. After lift-off process, the interdigital electrodes were formed and the rapid thermal annealing at 475 °C for 1 min was conducted. The room-temperature optical responsivities of β-Ga_2O_3 PD were measured by an Agilent B1500A semiconductor parameter analyzer and ultraviolet light emitting diodes (UV LEDs) emitting 254 and 356 nm light. Spectral-response measurements at −1 V bias were performed by spectrometer equipped with Xe light source. An Si detector was tested for calibrating the spectrum response.

3. Results and discussion

The crystal property of β-Ga_2O_3 films was measured by XRD with ω–2θ scan mode. Fig. 1(a) shows the XRD spectra of Ga_2O_3 films with varying T_B from 650 to 800 °C. Three diffraction peaks at 18.9°, 38.3°, and 58.9° exhibit well-defined (−201), (−402), and (−603) planes of β-Ga_2O_3, respectively, indicating the (−201) oriented single crystal growth. This is owing to the similar oxygen atomic arrangement of (−201) plane β-Ga_2O_3 and c-sapphire. The diffraction peak intensity of β-Ga_2O_3 film with T_B = 650 °C is weak, which is likely due to the low migration rate of adsorbed atoms on substrate surface at low growth temperature. When increasing T_B, the crystalline quality of epitaxial layer is improved and the intensity of diffraction peak increases. Figs. 1(b) gives the ω scanning diffraction pattern of (−402) β-Ga_2O_3 planes and the FWHM values at different T_B. The FWHM values of films prepared at different T_B of 650, 700, 750, and 800 °C are 3.3°, 0.66°, 0.67°, and 0.93°, respectively. The β-Ga_2O_3 films in this work grown by two-step method have much lower FWHM values, compared with previously reported β-Ga_2O_3 films on sapphire[19–21]. The sample grown at T_B = 700 °C shows the lowest FWHM value, indicating the optimized buffer layer growth condition is found.

AFM was performed to further investigate the surface morphology of films deposited on c-sapphire with different T_B. In Fig. 2(a), four samples were examined by AFM scan on a 5 × 5 μm scan area. The surfaces of four samples all show regular pseudo-hexagon structures, which are generated by the in-plane rotational domains and attributed to the symmetries of c-sapphire and β-Ga_2O_3[22, 23]. The film with T_B = 650 °C presents the sparsest texture, which is owing to the low mobility of crystal nuclei during the initial growth and lead to sparse grains difficult to merge. The sample with T_B = 700 °C exhibits the densest texture, which is likely attributed to the moderate crystal nuclei mobility at this temperature. As the temperature rises (with T_B = 750 or 800 °C), the atom mobility increases, leading to the deposition of large crystal nuclei on substrates and resulting in rough surfaces. Fig. 2(b) shows the surface roughness of four samples, which are 19, 6.8, 7.5, and 12 nm, respectively. Fig. 2(c) gives the variation in film thickness with different T_B and inset shows the cross-sectional SEM image of sample grown with T_B = 700 °C. The thickest β-Ga_2O_3 film of 1 μm is obtained when T_B = 700 °C. These results indicate that the buffer layer temperature at 700 °C is favorable for β-Ga_2O_3 film two-step growth.

The MSM solar-blind photodetector was fabricated on β-Ga_2O_3 film with T_B = 700 °C. Fig. 3(a) shows the optical microscope diagram of device. Fig. 3(b) reveals the three-dimensional structure diagram of the device. Fig. 3(c) shows the I–V characteristic curve of MSM photodetector. As revealed in figure, the dark current is 400 pA at 10 V bias. Moreover, the I–V curve increases only a little under 365 nm light (light intensity of 15 mW/cm^2), indicating that β-Ga_2O_3 film is not sensitive to 365 nm light. However, the I–V curve increases steeply under 254 nm light (light intensity of 1.9 mW/cm^2), suggesting strong light response characteristics and excellent solar-blind photoelectric characteristics.

The light/dark current rejection ratio (PDCR)[24], which is defined as $(I_{photo} - I_{dark})/I_{dark}$, achieves an extremely large value of more than 10^6, indicating that the device has extremely high sensitivity. The responsivity of MSM detector with effective illumination area about 4.55 × 10^4 μm^2 reaches 1422 A/W under 10 V bias. Detectivity (D^*) can be used to evaluate the ability to perceive the lowest detectable signal, which is related to background noise. Assuming that dark current is the main source of device noise, D^* can be expressed as: $RS^{1/2}/(2qI_{dark})^{1/2}$ [24]. The photodetection based on high-quality β-Ga_2O_3 film has a D^* of 2.5 × 10^{15} Jones under 254 nm light, indicating the ultra-high signal-to-noise ratio. The excellent wavelength selectivity and ultrahigh D^* indicate that high sensitivity solar-blind detector is achieved.

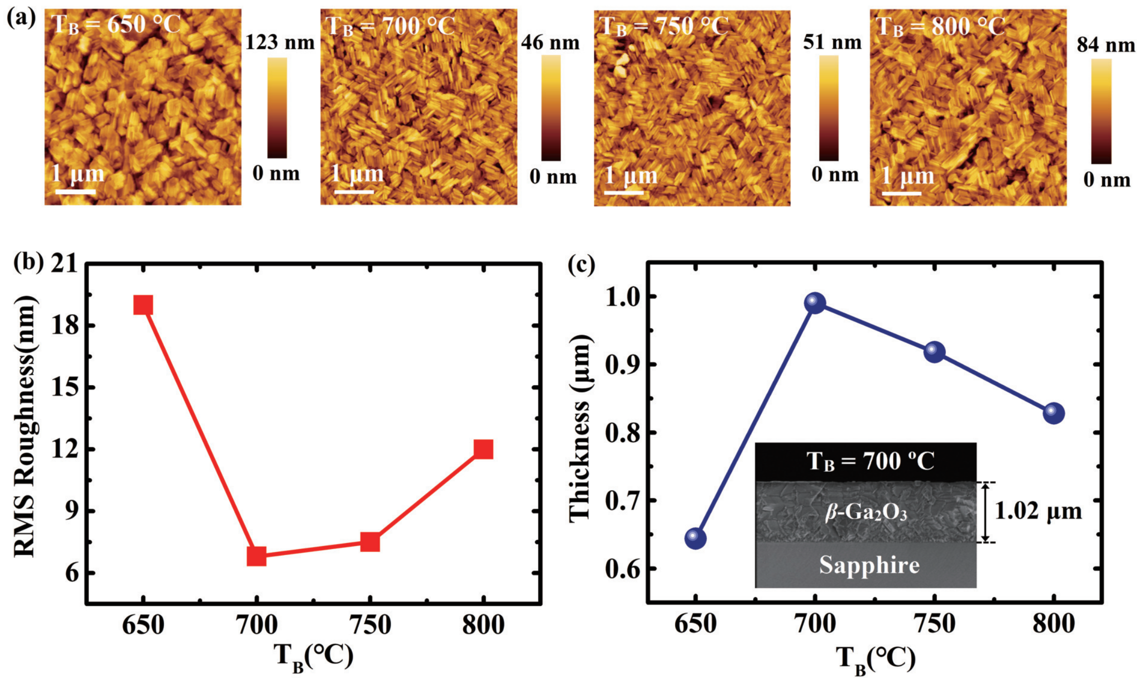

Fig. 2. (Color online) (a) AFM images and (b) RMS roughness of β-Ga_2O_3 films, (c) film thicknesses obtained from the cross-sectional SEM. Inset shows the cross-sectional image of film grown at T_B = 700 °C.

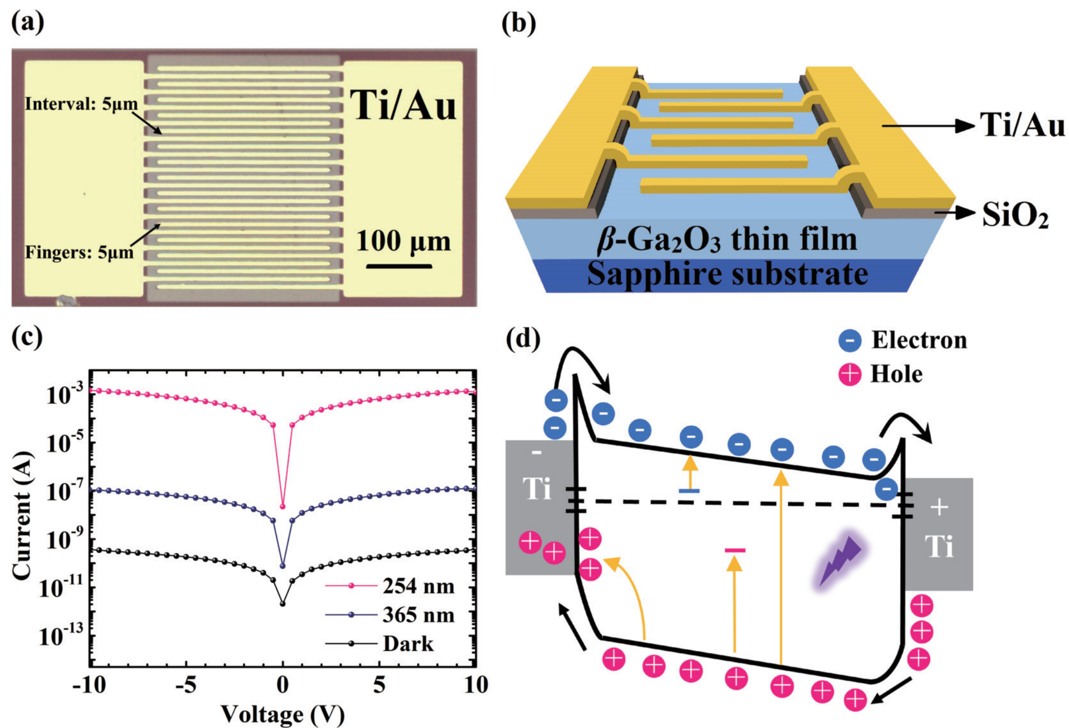

Fig. 3. (Color online) (a) The optical microscope diagram and (b) structural diagram of MSM photodetector. (c) The I–V characteristics of the MSM photodetector in the dark and illuminated by 254 and 365 nm. (d) The energy band diagram and photogenerated carriers of MSM β-Ga_2O_3 detector with external bias under 254 nm illumination.

Fig. 3(d) is energy band diagram and photogenerated carriers of the fabricated MSM photodetector under 254 nm light with external bias. It is determined from carrier transport mechanism that the optical response of photodetector is a complex process. Photogenerated carriers will be captured by deep-level defects so the lifetime is longer than transmission time. Moreover, electrons are quickly swept-out and reinjected to remain electric neutrality, generating multiple electrons and huge photoconductive gain.

The transient response of MSM photodetector under 254 nm wavelength illumination was tested by controlling the turning on and off for the light source, as shown in Fig. 4(a). When the light source is turned off, the current of the device decays quickly, showing a fast decay speed. In order to quantitatively analyze the response process of current rise and fall, the time responding characteristics are amplified as shown in Fig. 4(b). Then the double exponential relaxation equation is used to fit the I–T curve, and the equation is as follows[25]: I =

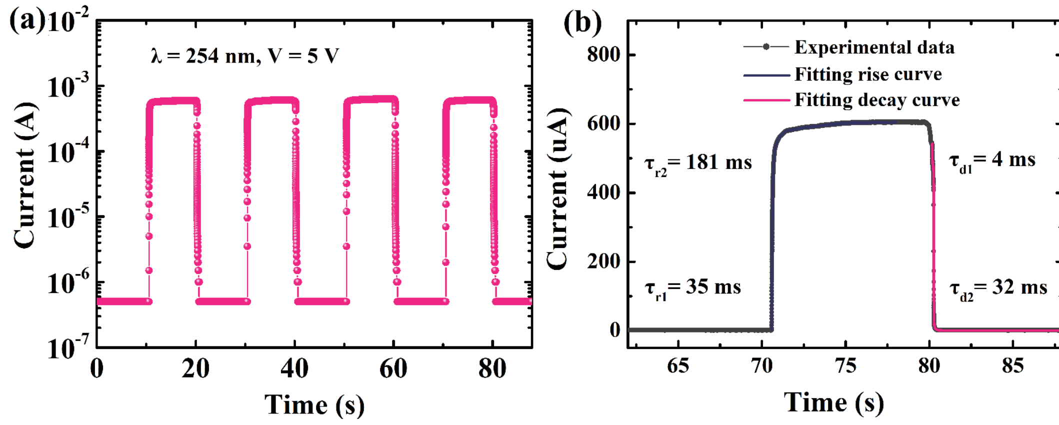

Fig. 4. (Color online) (a) The time-dependent photoresponse of the Ga_2O_3 photodetector with external bias to 254 nm illumination, (b) the experimental data and the fitting curve of rise and decay process.

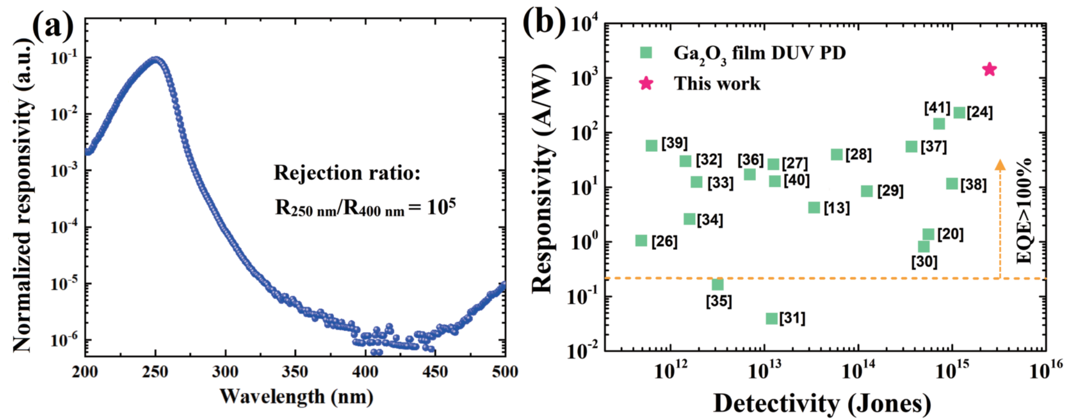

Fig. 5. (Color online) (a) The photocurrent spectrum of β-Ga_2O_3 MSM detector at −1 V bias in semilog scale, (b) the responsivity vs detectivity of different solar-blind photodetectors[32–41].

$I_0 + A\exp(-t/\tau_1) + B\exp(-t/\tau_2)$, where I_0 is the photocurrent in the stable state, t is the time, A and B are constants, τ_1 and τ_2 denote two relaxation time constants. τ_1 reflects the rapid change of carrier concentration after the light source is turned on or off, and τ_2 is caused by carrier capture/release due to defects in β-Ga_2O_3 film. By fitting the photoelectric response curve, the time constants of rising edge are τ_{r1} = 35 ms and τ_{r2} = 181 ms. The decay time constants of falling edge (i.e. the decay time) are: τ_{d1} = 4 ms and τ_{d2} = 32 ms. These values are faster than Ga_2O_3 photodetectors reported before[24, 26–29]. Furthermore, such fast decay speed (τ_d = 36 ms) indicates weak persistent photoconductivity (PPC) behavior, which can be caused by the reduced defect density in two-step growth film[29].

Fig. 5(a) shows the spectral response of the β-Ga_2O_3 MSM photodetector @−1 V and the peak wavelength is located at ~250 nm (4.97 eV), which is close to the bandgap of β-Ga_2O_3. The increased responsivity after 400 nm wavelength may be due to the absorption corresponding to the defect energy level. The rejection ratio ($R_{250\ nm}/R_{400\ nm}$) of 10^5 shows a higher response wavelength selectivity than previously reported results[30, 31]. Excellent solar-blind ultraviolet detection characteristic demonstrates the high-quality film and strong interband excited transitions. Fig. 5(b) gives the responsivities and detectivities of the reported deep ultraviolet photodetectors based on Ga_2O_3 films. It showed clearly that the obtained solar-blind photodetector displays the better comprehensive performance. In addition, the actual responsivity (R_{actual}) is given by $R_{actual} = GR_{ideal} = G(\eta q/h\nu)$[6], where η is EQE and ideal responsivity (R_{ideal}) is β-Ga_2O_3 photodetector detected at 254 nm. Assuming η is 100% and R_{ideal} is 0.2 A/W, the gain (G) is equal to 7110 indicating huge internal gain in β-Ga_2O_3 photodetector.

4. Conclusion

In summary, a Ga_2O_3 buffer layer is introduced to reduce lattice-mismatch-related defects using MOCVD method. It is found that T_B will obviously affect crystalline quality and the optimized buffer layer temperature for two step method is 700 °C. The MSM solar-blind photodetector shows ultrahigh responsivity of 1422 A/W and superior detectivity of 2.5× 10^{15} Jones. Besides, a notable rejection ratio ($R_{250\ nm}/R_{400\ nm}$) of 10^5 is realized, indicating the excellent wavelength selectivity. A remarkable gain of 7110 further demonstrates high internal gain in the β-Ga_2O_3 MSM photodetector. The outstanding photodetector performance suggests that two-step growth method has great potential for preparing high-quality β-Ga_2O_3 film on sapphire.

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