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Single-crystalline cubic MgZnO films and their application in deep-ultraviolet optoelectronic devices

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By employing a relatively low growth temperature and oxygen-rich conditions, single-crystalline cubic MgZnO films were prepared. A solar-blind deep ultraviolet (DUV) photodetector was finished on the MgZnO film. The maximum responsivity of the photodetector is 396 mA/W at 10 V bias, which is almost three orders of magnitude larger than the highest value ever reported in MgZnO-based solar-blind photodetectors. The dark current density is 1.5×10^{-11} A/cm^2, comparable with the smallest value ever reported in solar-blind photodetectors. The improved performance reveals that the single-crystalline cubic MgZnO films have great potential applications in DUV optoelectronic devices. © *2009 American Institute of Physics*. [doi:10.1063/1.3238571]

Optoelectronic devices operating in deep ultraviolet (DUV) region (with wavelength shorter than 300 nm) have attracted much attention in recent years for their potential applications in flame sensing, chemical/biological agents detection, convert communications, missile plume sensing, air and water purification, etc.[1–5] MgZnO, with a tunable band gap in the range of 3.3–7.8 eV, is a promising candidate for optoelectronic devices that covers a broad portion of the DUV spectrum.[6–12] Choopun *et al.*[13] have reported the wide band gap (5.0–6.0 eV) cubic MgZnO film. Recently, our group fabricated MgZnO films with their band gap in the range of 4.77–5.64 eV, and photodetectors with their cutoff wavelengths covering the whole solar-blind region (220–280 nm) have been obtained.[14] It is widely accepted that single-crystalline film is fundamentally important for high performance optoelectronic devices. However, since MgO prefers to crystallize in cubic rocksalt structure, while ZnO in hexagonal wurtzite structure, phase-separation will occur in MgZnO at a certain Mg content, which hinders drastically the realization of single-crystalline cubic MgZnO films.[8,13,15–17] No report on the realization of single-crystalline cubic MgZnO films can be found up to now.

In this letter, by employing a relatively low growth temperature and oxygen-rich conditions, single-crystalline cubic MgZnO films were prepared in metal-organic chemical vapor deposition (MOCVD) technique. To confirm the usefulness of the single-crystalline MgZnO films in high-performance DUV optoelectronic devices, a solar-blind DUV photodetector was fabricated based on the MgZnO films. The photodetector shows significantly smaller dark current and larger responsivity than the previous reported corresponding values, which proves the potential of single-crystalline MgZnO films in the DUV optoelectronic devices.

The MgZnO films were deposited on sapphire in a MOCVD system. Dimethyl dicyclopentadienyl magnesium (MCp2Mg), diethyl zinc (DEZn), and high pure oxygen (O_2) were employed as the precursors, and nitrogen as a carrier gas to lead the precursors into the growth chamber. The sapphire substrate was degreased in organic reagents and then etched in hot $H_3PO_4:H_2SO_4=1:3$ solution for 15 min. After that, the substrate was rinsed in deionized water, and blown dry with high-purity nitrogen before being loaded into the growth chamber. The deposition temperature was kept at 450 °C and the chamber pressure at 150 Torr during the growth process. The flow rate of MCp2Mg was fixed at 18 μmol/min, DEZn at 3 μmol/min, and O_2 at 0.07 mol/min providing a II/VI ratio of about 1:300. In this way, MgZnO films with a thickness of about 700 nm were obtained.

The transmission spectrum of the MgZnO films was recorded using a Shimadzu UV-3101PC scanning spectrophotometer. The crystal structure was studied by a Bruker D8 GADDS x-ray diffractometer (XRD) using Cu-Kα radiation with an area detector. The current-voltage (*I-V*) characteristic of the photodetectors fabricated from the MgZnO film was measured by a semiconductor parameter analyzer (Keithely 2200). The photoresponse of the photodetector was measured using a deuteron and xenon mixed light source and a Spex scanning monochromator.

The composition of the films determined by energy-dispersive x-ray spectroscopy is $Mg_{0.54}Zn_{0.46}O$. A typical transmission spectrum of the MgZnO films is shown in Fig. 1. The films show a high transmission of around 90% in the visible range, while they have a very sharp transmission edge at around 250 nm, which indicates the high optical quality of the films. The transmission edge lies in the DUV solar-blind spectrum range, suggesting that optoelectronic devices operating in the DUV region can be expected from the MgZnO films.

The structural characterizations of the MgZnO films were assessed by XRD, as shown in Fig. 2(a). Only two strong peaks at 36.62° and 77.68° can be observed in the θ-2θ XRD pattern besides the peak from the substrate, which can be indexed to the diffraction from cubic MgZnO (111) and (222) facets, respectively. The XRD data reveal that the films are crystallized in cubic structure with (111) preferred

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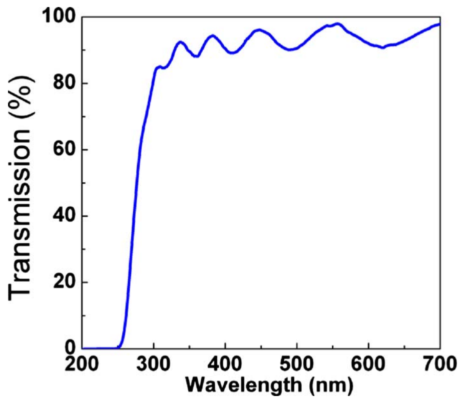

FIG. 1. (Color online) Transmission spectrum of the cubic MgZnO films.

orientation. X-ray rocking curve (XRC) of the (111) facet of the films shows a good symmetric Gaussian lineshape, and the full width at half maximum of the curve is 848 arcsec, as shown in Fig. 2(b). To determine the in-plane orientation of the MgZnO films, double-crystal XRD phi-scan was carried out at $\chi=35.2°$ [which is the angle between the (111) and (110) planes in a cubic system] from 0° to 360°. The scan yields three sharp peaks with 120° apart, as shown in Fig. 2(c). The phi-scan results indicate the threefold symmetry of the film, which is in good agreement with the XRD results that the film has (111) preferred orientation. Two-dimensional XRD (2D-XRD) was also used to explore the in-plane atom arrangement of the MgZnO films. One full diffraction ring in the 2D-XRD symbolizes the random arrangement of the in-plane atoms or molecules in the film. Only part of the diffraction ring will be obtained for a highly texture film. An ideal 2D-XRD image for a single-crystalline film was a focused point. The 2D-XRD image of the MgZnO films was shown in Fig. 2(d). The image of the film does not show any diffraction rings but gives a focused dot, which symbolizes the uniform in-plane atom alignment and structural isotropy of the film. The above results prove that the

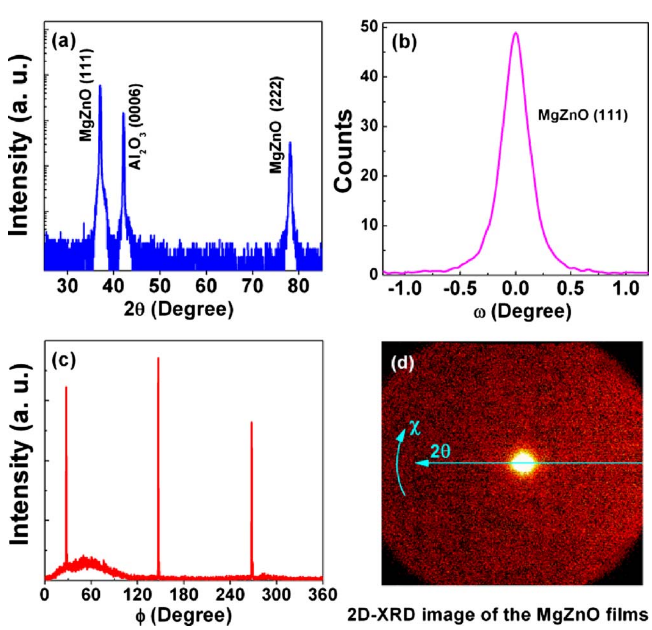

FIG. 2. (Color online) (a) θ-2θ XRD spectrum of the MgZnO films. (b) XRC of the MgZnO (111) reflection. (c) The phi-scan of the (110) reflection of the films. (d) 2D-XRD image of the MgZnO films.

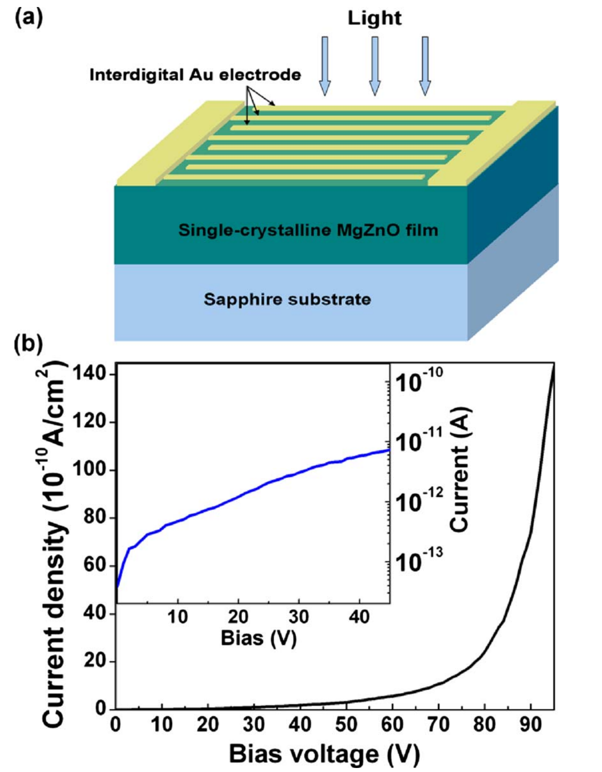

FIG. 3. (Color online) (a) Schematic illustration of the photodetector fabricated from the single-crystalline cubic MgZnO films. (b) Current density vs bias voltage of the MgZnO photodetector measured in dark, and the inset is the logarithmic *I-V* curve of the photodetector.

MgZnO films are single-crystallized with cubic structure. It is noteworthy that no report on single-crystalline cubic MgZnO films has been demonstrated before.

The formation of single-crystalline MgZnO films can be attributed to the following two factors: First, the relatively low growth temperature employed (450 °C) in the growth process is helpful to suppress phase-separation that is frequently observed in MgZnO alloys.[14] Thus, single phased films are obtained. Second, much redundant oxygen is introduced into the chamber during the growth process (the VI/II ratio is 300), which can suppress the formation of common intrinsic defects in ZnO-based materials, such as oxygen vacancies and interstitial zinc, thus may help to obtain MgZnO films with high crystalline quality.[18]

To explore the usefulness of the single-crystalline cubic MgZnO films in high-performance DUV optoelectronic devices, a photodetector was fabricated from the MgZnO films, and the structure of the device is schematically shown in Fig. 3(a). It consists of a 700 nm MgZnO film on sapphire substrate, and interdigital Au electrodes. The interdigital fingers was achieved via a photolithography and wet etching procedure. The finger was 3 mm in length and 2 μm in width, and the spacing between the fingers is 5 μm. A typical *I-V* curve of the MgZnO photodetector is shown in Fig. 3(b). The photodetector exhibits a dark current of about 0.45 pA at 10 V bias, which corresponds to a dark current density of about 1.5×10^{-11} A/cm^2 considering the effective area of the photodetector is about 3.0 mm^2. This value is comparable to the smallest dark current density value ever reported in solar-blind photodetectors (1.6×10^{-11} A/cm^2, which was realized in a AlGaN solar-blind photodetector).[19]

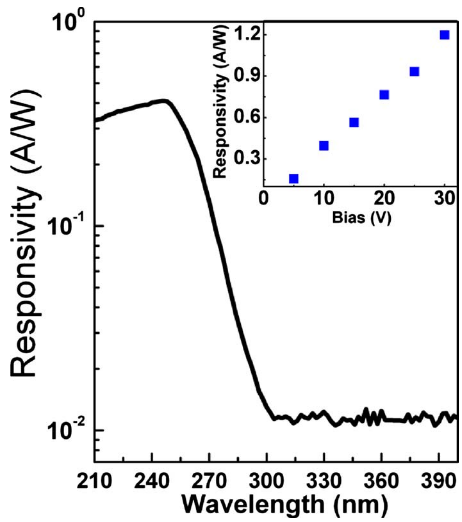

FIG. 4. (Color online) The photoresponse of the MgZnO photodetector at 10 V bias. The inset shows the dependence of the maximum responsivity of the photodetector on the bias applied.

The response spectrum of the photodetector is shown in Fig. 4. The photodetector shows a peak response at 246 nm, which is in reasonable agreement with the transmission edge of the MgZnO film (250 nm) shown in Fig. 1. The maximum responsivity is about 396 mA/W at 10 V bias, which is more than three orders of magnitude larger than the largest value ever reported in MgZnO-based DUV solar-blind photodetectors (0.1 mA/W).[14] The cutoff response wavelength of the photodetector is located at 265 nm, and the peak responsivity increases linearly with the bias applied in the investigated range, as shown in the inset of Fig. 4.

The improved performance of the DUV solar-blind photodetector can be attributed to the enhanced quality of the single-crystalline MgZnO films. It is accepted that the structural defects, such as grain boundaries and threading dislocations, will be greatly suppressed in the single-crystalline MgZnO films, thus less photogenerated carriers will be quenched by the defects. Consequently, more carriers can be collected by the interdigital electrodes, and the responsivity of the photodetector is improved. As for the relatively low dark current, thanks to the enhanced structural integrity of the MgZnO films, fewer intrinsic defects, such as oxygen vacancies and interstitial zinc, will be resulted, thus a reduced residual electron concentration can be obtained. As a result, the dark current of the photodetector is lowered.

In conclusion, single-crystalline cubic MgZnO films were achieved in an oxygen-rich ambient at relatively low temperature. To explore the usefulness of the films in DUV optoelectronic devices, a solar-blind photodetector was fabricated from the MgZnO films, and the responsivity of the photodetector is more than three orders of magnitude larger than the largest value ever reported in MgZnO-based solar-blind photodetectors. The dark current density of the photodetector is comparable to the smallest value ever reported in DUV solar-blind photodetectors. The enhanced performance of the photodetector reveals that the single-crystalline cubic MgZnO films have great promising applications in DUV optoelectronic devices, and the results reported in this letter thus address a significant step toward such applications.

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