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The effect of thermal annealing on $(In_2O_3)_{0.75}(Ga_2O_3)_{0.1}(ZnO)_{0.15}$ thin films with high mobility

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A B S T R A C T

We fabricated a series of $(In_2O_3)_{0.75}(Ga_2O_3)_{0.1}(ZnO)_{0.15}$ thin films using the pulsed laser deposition (PLD) method at room temperature under same oxygen pressure, and annealed these films at different temperatures from RT to 300 °C under vacuum. The effect of thermal annealing on the properties of In–Ga–Zn–O (IGZO) films was studied. The structural, optical and electrical properties of the films were measured by various diagnosis tools. Surface morphology of all IGZO thin films was examined by Atomic Force Microscopy (AFM). X-ray diffraction (XRD) patterns showed that the mixed crystalline and amorphous phases appear in the as-grown film and in the films annealed temperatures from 100 °C to 300 °C. The maximum carrier mobility was 32 $cm^2/(V\ s)$ in the film annealed at 300 °C. The transmission spectrum of film annealed at 300 °C shows the better light transmission and narrower band gap. The IGZO thin films with high mobility and transparency are highly desirable for the fabrication of flat panel display devices.

1. Introduction

Thin film transistors (TFTs) based on transparent oxide semiconductors (TOS), such as zinc oxide (ZnO) [1], indium tin oxide (ITO) [2], and other oxide compounds, have received considerable attention in the last few years because of both their good transparency, low processing temperature and large carrier mobility. In–Ga–Zn–O (IGZO) are expected to be the channel material of TFTs in next-generation flat-panel displays because IGZO TFTs satisfy most of the requirements for organic light-emitting-diode displays, large and fast liquid crystal and three-dimensional (3D) displays, which cannot be met using conventional silicon and organic TFTs. For such applications, the preparation of high quality In–Ga–Zn–O optical thin films with large electron mobility is an essential part of the channel material of TFTs. It is easy to obtain a-IGZO (In:Ga:Zn = 1:1:1) film with Hall mobility above 10 $cm^2\ V^{-1}\ s^{-1}$ [3], but further improvement of the mobility is difficult. Thus amorphous In–Ga–Zn–O thin films with high mobility and high transparency have been the subject of studies for many years.

In the paper, we prepared In–Ga–Zn–O films using pulsed laser deposition (PLD), and annealed the films at different temperatures. We also investigated the evolution of the structural, electrical and optical properties of these films as a function of annealing temperature. As a result, we found the coexistence of the amorphous and crystalline phase in the In–Ga–Zn–O films. It is also interesting to note that the annealed In–Ga–Zn–O thin films exhibit a significant enhancement of Hall mobility. The mechanism of the change of the electrical characteristics in In–Ga–Zn–O thin films induced by thermal annealing was explored.

2. Experimental procedure

A $(In_2O_3)_{0.75}(Ga_2O_3)_{0.1}(ZnO)_{0.15}$ ceramic target with a diameter of 30 mm was prepared from high purity powders at 1200 °C using the conventional solid state reaction method. Glass wafer with a size of $10 \times 10\ mm^2$ were placed at 45 mm away from the target. Prior to deposition, the chamber was evacuated to 3.3×10^{-4} Pa, and then the substrates were irradiated by a 355 nm beam from a frequency-tripled Nd: YAG laser (GCR-170, Spectra-Physics) for 60 min to improve adhesion of the IGZO film to the substrate. The target was then ablated at room temperature under 3 Pa oxygen pressure, and the film with ~300 nm thick was deposited by laser with a pulse duration of 10 ns, a repetition rate of 10 Hz and a pulse energy of 30 mJ. Following film deposition, the films were in-site annealed at different temperatures of 100 °C, 200 °C and 300 °C for 30 min, and then cooled to room temperature at a rate of 1 °C/min [4].

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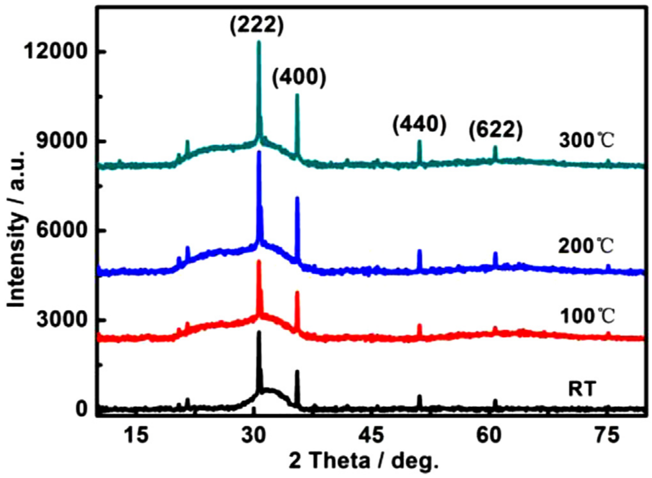

Fig. 1. (a) Variation of XRD patterns of $(In_2O_3)_{0.75}(Ga_2O_3)_{0.1}(ZnO)_{0.15}$ thin films with different annealing temperatures in vacuum.

The amorphicity or crystallinity of the films was checked by X-ray diffraction (XRD, BRUKER D8 ADVANCE) using a conventional x-ray diffractometer in a 2θ scan. The optical transmittance and optical band gap of the IGZO films were measured by UV–VIS spectrophotometer (UV 1900). The electrical properties were examined using a Hall-effect measurement system (Eastchanging ET 9000). The thickness of the films was measured by surface roughness meter (Veeco Dektak 150). The images of surface morphology were detected by atomic force microscopy (Veeco Multimode). All of the measurements were carried out at room temperature.

3. Results and discussion

Fig. 1 shows the X-ray diffraction patterns of the as-grown and annealed $(In_2O_3)_{0.75}(Ga_2O_3)_{0.1}(ZnO)_{0.15}$ thin films. All XRD spectra of the films contain some sharp peaks as well as a broad band. The sharp peaks in the films correspond to the cubic structure of polycrystalline In_2O_3 (222), (400), (440) and (622) as marked in Fig. 1. With increasing annealing temperature from 100 °C to 300 °C, the diffraction intensity increases gradually to the maximum. On the other hand, the broad band indicates that amorphous structure exists in the as-grown and annealed $(In_2O_3)_{0.75}(Ga_2O_3)_{0.15}(ZnO)_{0.05}$ thin films. Analysis of broad band indicates that there are two broad peaks centred at 23.86° and 31.18° that correspond to glass substrate and $(In_2O_3)_{0.75}(Ga_2O_3)_{0.15}(ZnO)_{0.05}$ film, respectively. Therefore X-ray diffraction (XRD) patterns reveal the coexistence of crystalline and amorphous phases in all the films.

The grain size D of the films can be calculated using the Scherrer's formula: $D = 0.9\lambda/B\cos\theta$. where $\lambda = 0.1540598$ nm and B is the measured full width at half maximum (FWHM) of the diffraction peak. As shown in Fig. 2(a), FWHM of the (222) peak in the films decreases from 0.175° to 0.14° with increasing annealing temperature from room temperature to 300 °C. The grain size of the films therefore were estimated to increases from 47.57 to 58.14 nm with increasing annealing temperatures from RT to 300 °C.

Fig. 3(a–d) display surface morphologies of the In–Ga–Zn–O films (5 μm–5 μm scanning area) annealed at RT, 100 °C, 200 °C and 300 °C, respectively. We choose this annealing temperature range because it fits real conditions of industrial production. The film annealed at more than 300 °C tends to deteriorate surface morphology and increase the surface roughness. The results showed that smooth surfaces could be obtained in the films annealed at 100 °C–200 °C. The root mean-square (RMS) roughness was 55.25 nm, 51.67 nm, 59.80 nm, and 125.16 nm for the thin films annealed at RT, 100 °C, 200 °C and 300 °C, respectively.

Fig. 4(a) shows the optical transmission spectra of $(In_2O_3)_{0.75}(Ga_2O_3)_{0.1}(ZnO)_{0.15}$ thin films annealed at different temperature. During the transmittance measurements, the effect of reflection and absorption of quartz glass substrate was eliminated by using amorphous quartz substrate as a reference. The average optical transmittance of the a-IGZO films at annealed at 300 °C is higher than 85% in visible light wavelength.

In a direct transition semiconductor, optical transmittance (T) and absorption coefficient (α) are correlated, as follows: $(\alpha h\nu)^2 = \beta (h\nu - E_g)$, where h is the Planck's constant, β is the energy-independent constant, and ν is the frequency of the incident photon. The optical band gap (E_g) can be obtained by extrapolating the straight-line portion of the ($\alpha h\nu$) [2] vs. $h\nu$ plot to the energy axis [5]. Fig. 4(b) shows that the optical band gap estimated from the optical transmission spectra of the films. It is evident that the band gap of $(In_2O_3)_x(Ga_2O_3)_y(ZnO)_{1-x-y}$ thin films is a correlated with the annealing temperature, as shown in the inset of Fig. 4(b).

The electrical properties of the as-deposited InGaZnO films annealed at temperature were evaluated by Hall effect measurement system under van der Pauw configuration [6,7], as shown in Fig. 5. In the present cases, all of the samples were found to be of the n-type. The results show that the Hall mobility increases and the resistivity decreases as the annealing temperature increases.

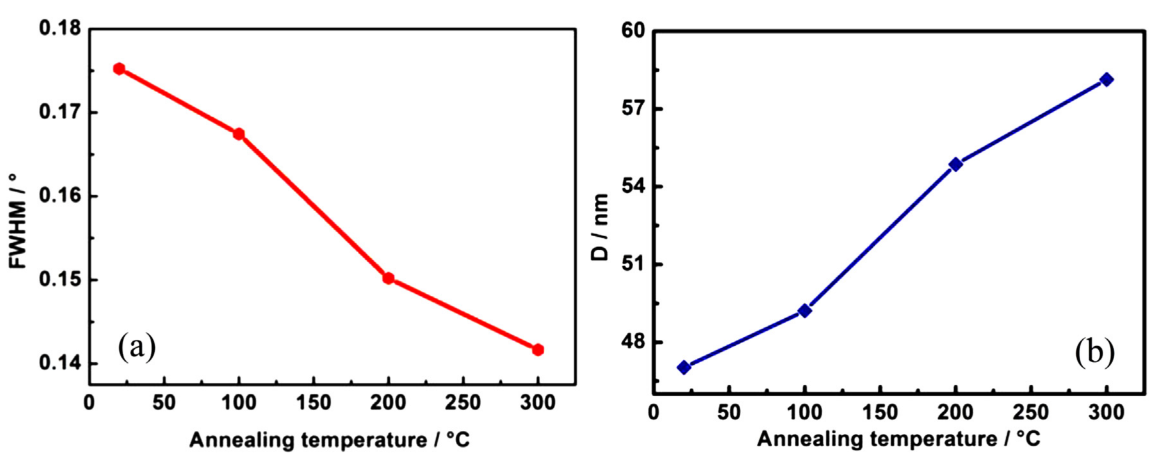

Fig. 2. (a) The full width at half maximum (FWHM) as function of annealed temperature for $(In_2O_3)_{0.75}(Ga_2O_3)_{0.1}(ZnO)_{0.15}$ thin films. (b) The grain size D function of annealed temperature.