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Oxygen incorporation in ZnTe thin films grown by plasma-assisted pulsed laser deposition

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ABSTRACT

We studied oxygen incorporation into ZnTe thin films with nitrogen and oxygen plasma during a plasma-assisted pulsed laser deposition (PA-PLD). It was shown that ZnTe:O layer formed with oxygen plasma exhibits an enhancement of optical transparency in visible spectral region due to the formation of amorphous TeO_x. Especially, the ZnTe:NO deposited by PA-PLD under nitrogen and oxygen partial pressures with N_2:O_2 of 10:3 sccm showed p-type semiconducting characteristics and the formation of intermediate band at about 0.5–0.8 eV below the ZnTe band edge. These results for oxygen incorporation in ZnTe thin film such as the enhancement of optical transparency in visible spectral region and the intermediate band formation will be useful for optoelectronic devices or intermediate band solar cells.
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1. Introduction

Zinc telluride (ZnTe) as one of II–VI family has a cubic crystal structure and a direct energy bandgap of 2.4 eV at room temperature. It is relatively low cost semiconducting material and a high optical absorption coefficient [1,2]. The ZnTe can be easily doped and shows usually p-type conducting [3–6]. Therefore, it can be used for solar cells, for example, as a back-surface field layer and p-type semiconductor material for CdTe/ZnTe heterojunction structure [7]. However, the carrier controllability of the ZnTe is an important issue to solve defect in optoelectronic devices [8]. Alloys of ZnTe:O is also applicable to optoelectronics devices owing to the tunable gap energy of the ZnTe:O in the visible spectral region. It is normally regarded that the oxygen atom becomes a shallow acceptor in II–VI semiconductors. Wang and Burki et al. reported that the large optical gain appears from the intermediate emission band of the ZnTe:O, which is measured at 0.4–0.6 eV below than the conduction band [9,10]. It means that there are two photon operations by two different wavelengths light source at room-temperature. The intermediate band approach offers the attractive prospect to achieving high efficiency in a more simplistic single-junction solar cell device. The theoretical efficiency for optimized triple-junction solar cells with efficiencies of 63.8% and 67.0% under the blackbody and the air mass (AM) 1.5 illumination [11], respectively, which are comparable to the theoretical conversion efficiency limit of 63.2% with blackbody illumination [12] and 65.1% with AM 1.5 spectrum [13] for intermediate band solar cells.

In this study, ZnTe:N, ZnTe:O and ZnTe:NO thin films were grown on Al_2O_3 (0001) substrates by using the plasma-assisted pulsed laser deposition (PA-PLD) with nitrogen and oxygen gases. The PA-PLD enables early control of the oxygen and the nitrogen concentration of the deposited films because of the enhanced reactivity of the dissociation and ionization of molecular and excited species [14,15]. The oxygen incorporation effect in the PA-PLD grown ZnTe thin films was analyzed and discussed for application of optoelectronic devices or intermediate band solar cells.

2. Experimental procedure

ZnTe:N, ZnTe:O and ZnTe:NO thin films were deposited on Al_2O_3 (0001) substrates by the PA-PLD with N_2:O_2 plasma. A pulsed (10 Hz) Nd:YAG laser operating at a wavelength of 266 nm using a nonlinear fourth-harmonic generator was used to produce a plasma plume from an ablated a ZnTe target, whose density of laser energy was 100 mJ/cm^2. The laser spot was focused to be about 1 mm^2 on the surface of the ZnTe target using an optical lens. The base pressure of the chamber was kept at a pressure of approximately 10^{-6} Torr using a turbo molecular pump. The oxygen gas flow was varied from 0 to 10 sccm, while the nitrogen gas flow was fixed at 10 sccm. The mixed gas conditions for the six samples, A to F, were summarized in Table 1. The working pressure

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Table 1
Oxygen and nitrogen gas flows during the growth of ZnTe films, and their energy band gaps.

Sample name	N_2 gas (sccm)	O_2 gas (sccm)	Band gap (eV)	Growth condition
A	10	0	2.47	Gas ambient
B	10	0	2.95	Plasma
C	10	3	(1.61)2.45	
D	10	5	4.59	
E	10	7	4.79	
F	0	10	4.56	

and RF power during the PA-PLD process were 10^{-4} Torr and 20 W, respectively. The thicknesses of all samples were controlled to be approximately 200 nm under the same growth temperature of about 300 °C.

The structural, chemical and optical properties of the ZnTeO thin films were investigated to study the influence of oxygen incorporation in ZnTe. X-ray diffraction (XRD) θ–2θ scans were used to determine crystal structure and crystalline orientation. A scanning electron microscopy (SEM) was used to study for the cross sectional morphology of the samples. Relative concentrations of oxygen, nitrogen, and associated bonding configurations were determined using X-ray photoelectron spectroscopy (XPS). XPS was used to detect the presence of oxygen-associated bonding configurations. The optical transmittance of the films was measured in the wavelength range of 200–800 nm using an ultraviolet–visible (UV–Vis) spectrophotometer.

3. Results and discussion

Fig. 1(a) and (b) shows XRD patterns for the six samples of ZnTe:N, ZnTe:O and ZnTe:NO thin films, and SEM cross sectional image for the ZnTe:NO layer, respectively. The samples of ZnTe:N, ZnTe:O and ZnTe:NO thin films were grown under the N_2:O_2 mixed gas plasma with the various oxygen gas flow from 0 to 10 sccm at fixed the nitrogen gas flow at 10 sccm. The ZnTe:NO on the Al_2O_3 (0001) substrate were examined to determine crystalline registration, where a preference for the epitaxial ZnTe(111) grown on c-plane sapphire substrates was observed based on x-ray diffraction (θ–2θ) scans as shown in Fig. 1(a). The diffraction peak of 24.91° correspond to the zinc-blende structured ZnTe(111). The highest diffraction peak of ZnTe(111) in the ZnTe:N thin film (Sample A) grown under N_2 gas ambient were observed at a growth temperature of 300 °C, and then the crystallinity of ZnTe:NO thin films of the B, C, D and E samples were observed to worsen with an increase oxygen gas flow. The ZnTe:NO thin films deposited by the PA-PLD under high oxygen and nitrogen plasma exhibited weak diffraction peaks corresponding to polycrystalline zinc blende material, suggesting a low degree of crystallinity and/or amorphous material. On the other hand, the diffraction peak of ZnTe(111) was not detected in the ZnTe:O thin film of the sample F grown by using oxygen gas plasma only without nitrogen gas. Typical SEM cross sectional image of the ZnTe:NO thin film grown on sapphire substrate for the C sample C is shown in Fig. 1(b). Here, the film thicknesses of the sample were about 200 nm.

The optical transmittances and the optical absorption spectra of the ZnTe:NO samples grown under plasma-assisted and gas ambient at various conditions of N_2:O_2 gas flow are shown in Fig. 2(a) and (b), respectively. In the energy range of 1.6–6.0 eV, the absorption curves of the B, C, D and E samples were red-shifted and blue-shifted from that of the ZnTe:N sample B, respectively. For determining the bandgap of the ZnO film, the absorption coefficient (α) is obtained from transmittance data using the following equation:

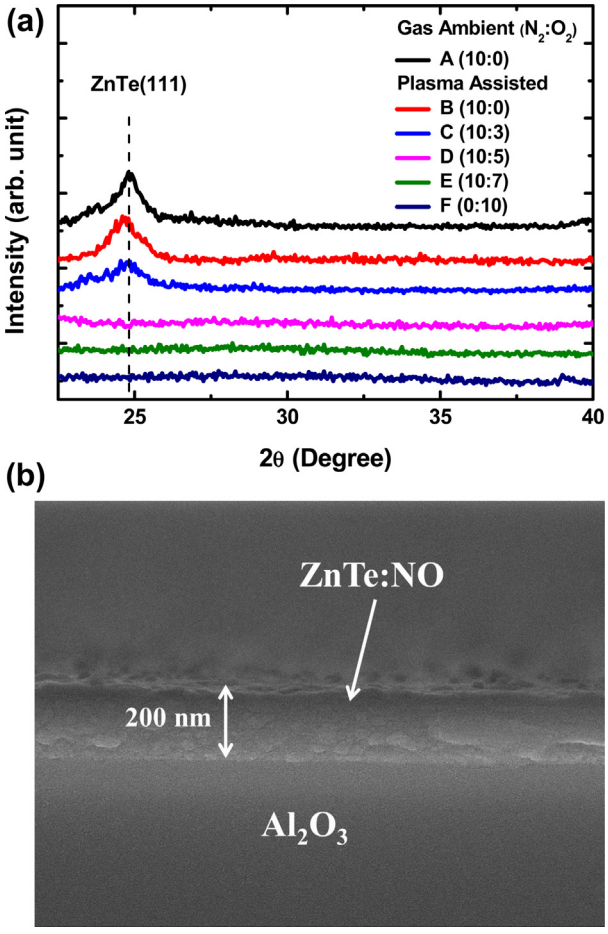

Fig. 1. (a) XRD patterns for ZnTe:N, ZnTe:O and ZnTe:NO thin films, and (b) SEM cross sectional image for the ZnTe:NO layer, respectively, which samples were grown under N_2:O_2 mixed gas plasma with various oxygen gas flow from 0 to 10 sccm at fixed nitrogen gas flow at 10 sccm.

$$\alpha = \frac{1}{d}\ln T \tag{1}$$

where d is the thickness of the film and T is the optical transmittance. The bandgap can be estimated using a well-known Tauc's relation [16].

$$\alpha(h\upsilon) = A(h\upsilon - Eg)^{\frac{1}{2}}, \tag{2}$$

where h is the Planck constant, υ is the frequency of the incident photon, and A is a constant. The estimated optical band gap of the ZnTe:NO samples is shown in Fig 2(c). Table 1 shows the optical energy band gap obtained from the absorption curves of ZnTe:N, ZnTe:O and ZnTe:NO films grown under N_2:O_2 plasma at various N_2 and O_2 gas flow from 0 to 10 sccm, respectively. Here, the optical absorbance of the A, B and C samples increased in the ranges from 2.5 to 2.9 eV as increase the oxygen gas flow. In the energy range of 1.6–2.0 eV, the absorption curves of the A and B samples showed a direct band-gap transition at 2.26 eV. The absorption spectra of sample C were red-shifted than the other A and B samples in the range of 1.6–2.0 eV. In general, the red-shifted was caused by the two emission bands because of the oxygen impurities and deep-level defects related to oxygen incorporation in thin films [17]. The oxygen incorporation of ZnTe thin films grown by PA-PLD produced an energy band structure at about 0.9 eV below the ZnTe band edge which indicated strong radiative properties

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