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Effects of cation compositions on the electronic properties and optical dispersion of indium zinc tin oxide thin films by electron spectroscopy

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ABSTRACT

The electronic properties and optical dispersion of indium zinc tin oxide (IZTO) films with different cation compositions were investigated by reflection electron energy loss spectroscopy (REELS). The REELS spectra of IZTO films revealed that the band gap varied with different Sn/Zn ratios and In content. The optical properties were examined with REELS data using Tougaard–Yubero model and the results were compared with the envelope of the transmission spectra obtained using a UV-spectrometer. The dispersion behavior of the refractive index from REELS results was studied in terms of the single-oscillator Wemple–DiDomenico model. The results showed that the different compositions of In/Zn/Sn caused a change in the dispersion parameters of IZTO thin films in contrast to the static values of refractive indices and dielectric constant which remained the same. Our work demonstrated that REELS is an efficient tool to study the optical properties of a material by obtaining the optical parameters.

1. Introduction

Indium zinc tin oxide (IZTO) thin films have applications as transparent conducting oxide (TCO) materials that serve as the active channel of thin-film transistors (TFTs). This is due to their high performance compared to conventional TFTs based on hydrogenated amorphous silicon (a-Si:H) having high mobility (>10 $cm^2 V^{-1} s^{-1}$), low gate sub-threshold swing, and high on-off current ratio (>10^6) [1–3]. Both indium tin oxide (ITO) and IZTO thin films have been employed as transparent electrodes for flat panel displays, touch panels, organic light emitting diodes, and solar cells because they satisfy the requirements for applications such as electrical resistivity of $\sim 10^{-4}\,\Omega$ cm and optical transmittance of 85% in the visible light region [4]. In comparison to ITO thin films, IZTO thin films are preferred because of their higher electrical conductivity, higher transparency, moderate chemical stability, higher work function, and low deposition temperature [5–7]. IZTO is also cost-effective because indium is pricy and doping ZnO into ITO can decrease the use of indium by 60% without deteriorating the electrical and optical properties [8,9].

Several studies have attempted to change the electrical, optical, and structural properties of IZTO thin films by using an annealing process and/or varying the composition of Zn and Sn. A recent study reported on IZTO thin films grown by RF magnetron sputtering with two different chemical compositions [10]. In this work, the In content was fixed to 60%, while the Sn content was varied between 15% and 25%. They found that the crystallinity of an IZTO thin film increases with a higher Sn content and exhibited much lower resistivity of $3.44 \times 10^{-4}\,\Omega$ cm when deposited at 400 °C. Saji and Jayaraj [11] made IZTO thin films by a co-sputtering method at room temperature (RT) with different composition ratios of Zn/In/Sn which varied from 0.05/0.56/0.39 to 0.5/0.27/0.23 and confirmed that a change in the compositions caused a change in the optical band gap which varied from 3.0 eV to 3.44 eV with increasing Zn content. They also showed that IZTO TFTs with a high performance can be realized at RT.

In this paper, we demonstrate that the electronic and the optical properties can be improved by using a lower content of In in IZTO thin films. We made two different kinds of IZTO samples with In contents of 20% and 13%, which has not been previously reported.

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We investigated the effect of different compositions of In/Zn/Sn on the energy band gap and optical properties using reflection electron energy loss spectroscopy (REELS) as well as the Tougaard–Yubero model.

In fact, REELS is a convenient technique to investigate the electronic structure of a material because the low-energy-loss region reflects the structure of the valence and the conduction electron bands [12]. The electronic structure can be obtained by analyzing the energy distribution of electrons reflected from the surface of a solid using a monoenergetic primary electron beam as in REELS [13].

The most conventional methods to measure the optical properties of transparent thin films are the envelope method using transmittance (or reflectance) spectra and the interferometric analysis method [14,15]. The interferometric method with two [16] or multiple beam interferometry and prism coupling technique [15] provide precise measurements of optical parameters, but to carry out the measurements complex tools and special experimental skills are needed [14]. On the other hand, the envelope method in transmittance spectra obtained by a UV-spectrometer uses a relatively simple procedure which depends on interference fringes. In the envelope method the optical parameters are determined by analysis of the interference fringes from transmittance spectra. However some films do not show interference fringes in the UV-spectroscopic measurement. REELS is an alternative technique to analyze optical parameters of thin films and when it is used in conjunction with the Tougaard-Yubero model (QUEELS-ε(k,ω)-REELS) it can provide information about the optical parameters of a thin film [13,17]. This method determines the dielectric function of the film, and from this, the optical parameters such as the refractive index, extinction coefficient, and transmission spectra can be easily determined. The validity of this method has been proven and applied to obtain optical parameters for ultrathin gate oxide films, dielectric materials, and transparent conducting oxides [18,19].

In our study, we demonstrated that the energy band gap was changed with lower In contents of 13% and 20%, and the optical properties were analyzed in terms of a single-oscillator method proposed by the Wemple and DiDomenico dispersion relation model [32].

2. Experimental

IZTO thin films were deposited on glass substrates at RT in argon mixed with oxygen gas ($Ar:O_2 = 15:85$) by RF magnetron sputtering with an RF power of 200 W. IZTO thin films were produced with composition ratios of In:Zn:Sn of 20:50:30 (denoted as IZTO-I), 20:56.7:23.3 (denoted as IZTO-II), and 13:60.2:26.8 (denoted as IZTO-III). The Sn/Zn ratios for IZTO-I, IZTO-II, and IZTO-III films are approximately 0.6, 0.42, and 0.45, respectively. Thus, the Sn/Zn ratios for IZTO-II and IZTO-III films are close to each other and those values are much lower than that of IZTO-I film. The thickness of all the thin films was 35 nm. All IZTO thin films were annealed at 350 °C for 1 hour in air. The REELS spectra were obtained using a VG ESCALAB 210 equipment. The REELS spectra were measured with primary electron energies of 1.0, 1.5, and 2.0 keV and the energy distribution of backscattered electrons were measured with a hemispherical electron energy analyzer operated at a constant analyzer pass energy of 20 eV. The electron incident and exit angles from the surface normal were 55° and 0°, respectively. The full width at half maximum (FWHM) of the elastic peak was 0.8 eV. The analysis of the crystal structure was carried out on a Phillips PW-1710Cu-Kα diffractometer (λ = 1541 Å) by varying the diffraction angle 2θ from 30° to 80° with an increment of 0.009°. The surface morphology including the roughness calculation of the films was performed by atomic force microscopy (AFM) from Nanofocus Inc. system with a scanning area of $2 \times 2\ \mu m^2$. The transmittance spectra of the IZTO thin films were measured by Genesys 6 model from Thermo Electron Corporation in the wavelength range of 300–1000 nm at RT at increments of 0.1 nm.

3. Results and discussions

Fig. 1 shows the REELS spectra and UV-spectroscopic results for the IZTO thin films. The results for annealed samples in Fig. 1 were

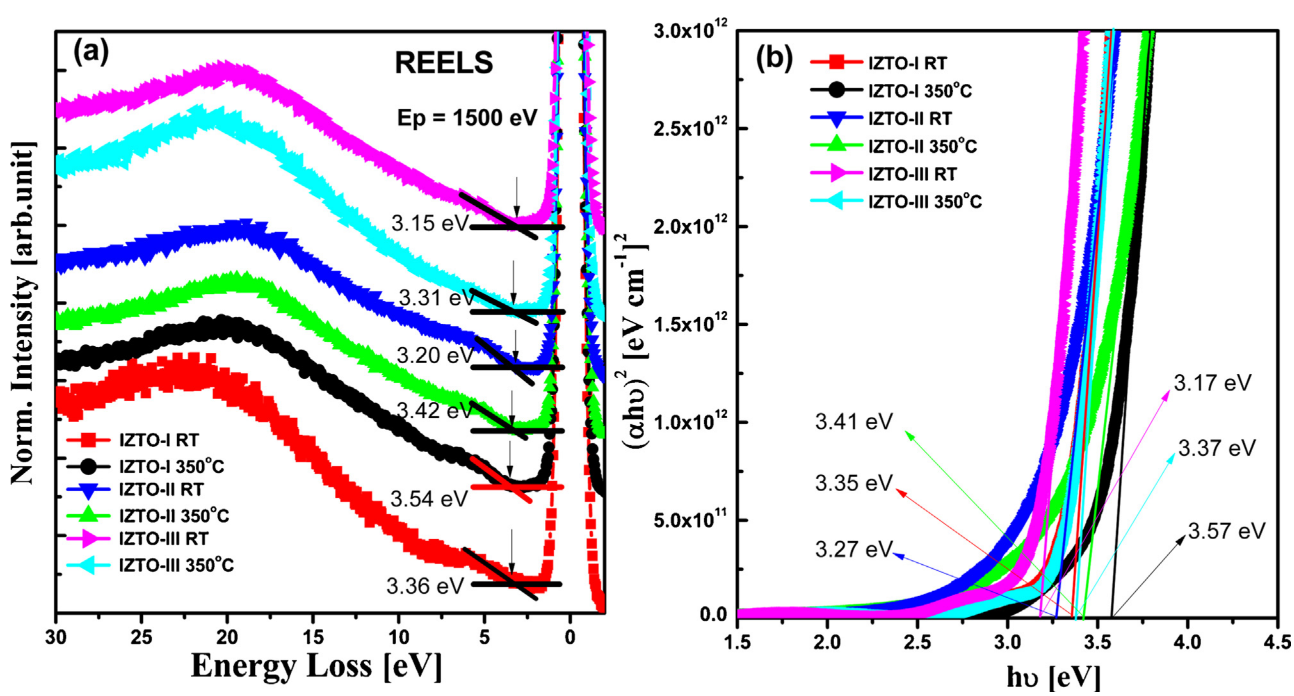

Fig. 1. (a) Reflection electron energy loss spectra with the primary energy of 1.5 keV and (b) plot of $(\alpha h\upsilon)^2$ versus $h\upsilon$ of IZTO thin films for different composition of In/Zn/Sn.