Indium-doped Mg$_{x}$Zn$_{1-x}$O films for ZnO-based heterojunction diodes.

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1. Introduction

ZnO is a semiconductor which has a wide band gap of 3.28 eV and a large exciton binding energy of 60 meV at room temperature, indicating a potential application for high-performance optical devices[1]. We have successfully achieved ZnO-based bandgap engineering by developing Mg$_{x}$Zn$_{1-x}$O and Zn$_{1-y}$Cd$_{y}$O material systems with the optical band gaps from 3.7 eV down to 1.8 eV, which are grown by remote-plasma-enhanced metalorganic chemical deposition (RPE-MOCVD)[2-4]. Recently, we have also demonstrated RGB full-color electro-luminescence emissions from ZnO-based heterojunction diodes [5], which provides a clear feasibility on ZnO-based material systems for optical applications. However, surface morphologies of the films grown by RPE-MOCVD as well as conventional MOCVD show relatively a tendency to be pillar-shaped, comparing with the films grown by molecular beam epitaxy [6].

In this paper, we describe structural improvements such as surface morphologies and crystallinity of indium doped Mg$_{x}$Zn$_{1-x}$O:In systems and green electroluminescences from ZnO-based double heterojunction (DH) system using Mg$_{x}$Zn$_{1-x}$O:In films.

2. Experimental details

Indium-doped Mg$_{x}$Zn$_{1-x}$O (Mg$_{x}$Zn$_{1-x}$O:In) and indium-doped ZnO (ZnO:In) films were prepared by RPE-MOCVD. Diethylyzinc (DEZn), bis-ethylcyclopentadienyl magnesium (EtCp$_2$Mg) and dimethyl cadmium (DMCd) were used as group-II sources and trimethylindium (TMIn) was used as group-III element basically serving n-type doping. Oxygen radicals as group-VI sources were generated from O$_2$ gas by a RF source with 13.56 MHz, and were introduced onto the heated substrates during the growth. Hydrogen was used as a carrier gas for metalorganic sources. Mg$_{x}$Zn$_{1-x}$O:In films and or ZnO:In films were grown on a-plane sapphire substrates. The crystallinity was characterized by X-ray diffraction (XRD) employing CuK$_\alpha$ line. Morphologies of the films were characterized by field-emission scanning electron microscopy (FE-SEM). The alloy composition was basically analyzed by atomic absorption spectroscopy. The electrical properties of ZnO-based double heterojunction systems discussed later were analyzed by the I-V measurement and electroluminescence spectra measurement.

3. Results and discussion

Figs. 1 (a) to (d) and Figs. 2 (a) to (d) show FE-SEM plan-views and corresponding cross-sectional images of the surface morphologies of Mg$_{0.05}$Zn$_{0.95}$O:In and ZnO:In with different amounts of TMIn, respectively. In the case of Mg$_{0.05}$Zn$_{0.95}$O:In films in Fig. 1, an increase in the grain size and the coalescence are seen along with an increment of the amount of TMIn, and the surface morphologies are clearly changed to become smooth. On the contrary, it seems that the grain sizes of the ZnO:In films shown in Fig. 2 basically becomes larger, but includes different grain size with an increment of TMIn. The difference of surface morphologies between Mg$_{0.05}$Zn$_{0.95}$O:In and ZnO:In may be possibly due to the migration enhancement effect on the magnesium atom.

Figs. 3 (a) and (b) shows dependency of XRD rocking curve full width half maximums (FWHMs) of Mg$_{0.05}$Zn$_{0.95}$O:In films and ZnO:In films as a function of [TMIn] mole fraction, respectively, indicating that Mg$_{0.05}$Zn$_{0.95}$O:In films had improved into smaller ones with an increase of the amount of TMIn and shown the value of 0.72$^\circ$ at TMIn of 3 %. In contast, the FWHMs of ZnO:In films does not show a clear dependency and relatively becomes larger than that of undoped ZnO. Comparing Mg$_{0.05}$Zn$_{0.95}$O:In at TMIn of 3 % with ZnO:In, the FWHMs have decreased by 40 %. The reason of the narrower improvement is possibly originated from the surface morphology change from to be pillar-shaped into the film-like as shown in Fig. 1. Figs. 4 shows dependency of optical band gap of both Mg$_{0.05}$Zn$_{0.95}$O:In films and ZnO:In films as a function of [TMIn] mole fraction, respectively. It is found that the optical band gaps change shows a tendency of red-shift with an increase of the amount of TMIn.

We have grown ZnO-based heterojunctions on p-4H-SiC substrate employing Mg$_{0.05}$Zn$_{0.95}$O:In film. Fig. 5 shows the schematic cross-section of the DH systems. Figs. 6 (a) and (b) show the I-V characteristics of the ZnO-based DH systems with indium-doped and undoped films, respectively. While a clear change of I-V characteristics is not found, the green EL emission from the DH system with the indium-doped film is enhanced by factor two, comparing with the undoped case as shown in Fig.7. Moreover, the EL spectra-FWHM has decreased by 13 % under an injection current of 100mA at room temperature. We are speculating that this result is mainly due to the improvement on the heterointerface between the active layer and the barrier layer by using Mg$_{0.05}$Zn$_{0.95}$O:In film, which are clearly found in the cross-sectional FE-SEM image of Fig. 8.

4. Conclusions

We have successfully improved surface morphology of Mg$_{0.05}$Zn$_{0.95}$O:In films from the pillar-like into the film-like with an increment of the amount of Indium and shown green strong electroluminescence emissions from ZnO-based heterojunctions.

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5. References


Fig. 1 FE-SEM surface morphologies of Mg$_{0.05}$Zn$_{0.95}$O:In versus [TMIn] mole ratio. The typical indium content for the case of 3.0% was calibrated by RBS measurement by 2.5%.

Fig. 2 FE-SEM surface morphologies of ZnO:In versus [TMIn] mole ratio.

Fig. 3 XRD rocking curve-FWHM of (a) Mg$_{0.05}$Zn$_{0.95}$O:In film and (b) ZnO:In films versus [TMIn] mole ratio.

Fig. 4 Optical band gap of (●) Mg$_{0.05}$Zn$_{0.95}$O:In film and (○) ZnO:In films versus [TMIn] mole ratio.

Fig. 5 The schematic crosssection of DH systems.

Fig. 6 I-V characteristics of DH systems (a) using Mg$_{0.05}$Zn$_{0.95}$O:In film and (b) undoped Mg$_{0.05}$Zn$_{0.95}$O films,respectively.

Fig. 7 (a) EL intensity versus injection current and (b) EL spectra under an injection current of 100 mA at room temperature.

Fig. 8 FE-SEM images of DH systems of (a) using Mg$_{0.05}$Zn$_{0.95}$O:In film and (b) undoped Mg$_{0.05}$Zn$_{0.95}$O films.