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To cite this article: K Saito et al 2008 J. Phys.: Conf. Ser. 100 042028

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Growth of undoped ZnMgTe layers by metalorganic vapour phase epitaxy

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Abstract. The ternary undoped Zn$_{1-x}$Mg$_x$Te layers have been grown on (100) ZnTe substrates by atmospheric pressure metalorganic vapour phase epitaxy (MOVPE) using dimethylzinc (DMZn), bis-methylcyclopentadienyl-magnesium ((MeCp)$_2$Mg) and diethyltelluride (DETe). The effects of the transport rates of DMZn and (MeCp)$_2$Mg upon the Mg composition and low temperature photoluminescence (PL) property of Zn$_{1-x}$Mg$_x$Te layer have been investigated. The PL spectra exhibit two distinctive luminescence bands, i.e. excitonic emission (band A) and donor-acceptor pair recombination emission (band B). The intensity ratio of the peak A to peak B increases with decreasing DMZn transport rate, indicating that the optical property of the layer is improved. The control of the Mg composition by (MeCp)$_2$Mg transport rate under VI/II ratios of one to two has an advantage in achieving both the controllability of Mg composition and good optical property of undoped Zn$_{1-x}$Mg$_x$Te layer grown by atmospheric pressure MOVPE.

1. Introduction

The ternary Zn$_{1-x}$Mg$_x$Te epitaxial layer is promising for application as cladding layer of ZnTe based pure green light-emitting devices, because of the wider band gap than ZnTe. However, there are only a few papers on Zn$_{1-x}$Mg$_x$Te epitaxial layer grown by metalorganic vapour phase epitaxy (MOVPE) [1-4], which is a potential growth technique for mass production. In our previous study [4], Zn$_{1-x}$Mg$_x$Te epitaxial layers have been successfully grown on (100) ZnTe substrates by atmospheric pressure MOVPE using dimethylzinc (DMZn), bis-methylcyclopentadienyl-magnesium ((MeCp)$_2$Mg) and diethyltelluride (DETe). It has been found that the Mg composition in the layer drastically increases with decreasing DETe transport rate or with increasing the total carrier gas flow rate, which may be associated with the reduction of the premature reaction between DETe and (MeCp)$_2$Mg. On the other hand, it has been also found that the Mg composition is not influenced significantly by DMZn transport rate at transport-limited mode. In order to obtain Zn$_{1-x}$Mg$_x$Te layers of good quality, it is necessary to investigate the relationship between the growth condition and optical property, but there are no reports on it. In this work, we studied the effect of the VI/II ratio of source transport rate upon the optical property by varying the transport rate of DMZn. Furthermore, under the obtained optimal VI/II ratio, the controllability of Mg composition and effect upon the optical property was examined for various transport rates of (MeCp)$_2$Mg.
2. Experimental procedure
The nominally undoped Zn$_{1-x}$Mg$_x$Te epitaxial layers were grown with atmospheric pressure MOVPE system with a vertical reactor in almost the same manner as was employed in the previous study [4]. Briefly, DMZn, (MeCp)$_2$Mg and DETe were used as source materials while hydrogen was employed as a carrier gas. The growth experiments have been carried out at substrate temperature of 380 °C. In order to investigate the effect of VI/II ratio of source transport rate upon the photoluminescence property, DMZn transport rate or (MeCp)$_2$Mg transport rate was varied, respectively, from 5 to 20 μmol/min (VI/II ratio: 0.45~1.4) or from 0 to 5 μmol/min (VI/II ratio: 1~2), while transport rate of DETe and total hydrogen flow rate were, respectively, fixed at 10 μmol/min and 800 sccm.

The Mg composition in Zn$_{1-x}$Mg$_x$Te layers was determined by energy dispersive X-ray (EDX) analysis. PL measurement was performed at 4.2 K using 457 nm Ar$^+$ laser or 405 nm blue-violet laser diode as an excitation light source.

3. Results and discussion
Figure 1 shows the dependence of DMZn transport rate on Mg composition in the Zn$_{1-x}$Mg$_x$Te layers. It is noteworthy that the Mg composition slightly increases with increasing DMZn transport rate in spite of the decrease in the pressure ratio of the Mg precursor to Zn one in gas phase which may compete to occupy the same lattice site. The similar dependence was also observed for the Zn$_{1-x}$Mg$_x$Te layers grown at a substrate temperature as high as 420 °C [4]. Figure 2 shows the low temperature PL spectra of Zn$_{1-x}$Mg$_x$Te layers for various transport rates of DMZn. The PL spectra are characterized by two distinctive luminescence bands denoted by A and B in the figure. Referring to the literatures [5,6], the A band is considered to be excitonic emission. For the B band, the peak shifts to shorter wavelength side as the excitation power increases, which is a characteristic of a donor-acceptor pair recombination emission (DAP) [7]. As shown in the inset of figure 2, the intensity ratio of the peak A to peak B increases with decreasing DMZn transport rate, indicating that the optical property of the...
layer is improved at high VI/II ratio. Asano et al. suggested that the impurity substituted into a Te site is responsible for the DAP [3]. The high VI/II ratio is found to be essential for improving the optical property of the undoped Zn$_{1-x}$Mg$_x$Te layer. However, such a condition attained by high transport rate of DETe or low transport rate of DMZn is an adverse condition for the incorporation of Mg into Zn$_{1-x}$Mg$_x$Te layer, since premature reaction between DETe and (MeCp)$_2$Mg is enhanced [4]. Therefore, the control of the Mg composition by (MeCp)$_2$Mg transport rate under a suitable VI/II ratio is expected as a promising growth condition for achieving both the controllability of Mg composition and relatively good optical property.

Figure 3 shows the Mg composition of Zn$_{1-x}$Mg$_x$Te layers for various transport rates of (MeCp)$_2$Mg, grown at a DMZn transport rate of 5 μmol/min. The Mg composition increases monotonically with increasing of transport rate of (MeCp)$_2$Mg. As the transport rate of (MeCp)$_2$Mg increases, the incorporation efficiency increases gradually. Thus, the transport rate of (MeCp)$_2$Mg is effective to control the Mg composition as well as the transport rate of DETe or total carrier gas flow rate [4]. Figure 4 shows the low temperature PL spectra of Zn$_{1-x}$Mg$_x$Te layers for various transport rate of (MeCp)$_2$Mg. In this measurement, 405 nm blue-violet laser diode was used as excitation light source to extend the measurable wavelength region. In each PL spectrum, no deep emission is appeared in the wavelength region between 600 to 700 nm (not shown). The PL spectrum of the Zn$_{1-x}$Mg$_x$Te layer grown at (MeCp)$_2$Mg transport rate of 0 μmol/min, i.e. ZnTe layer, composed of a free exciton emission (FE) at 520.5 nm and strong excitonic emission at 522 nm. In the PL spectra of $x > 0$, FE almost vanishes, excitonic emission (band A) broadens and weak donor-acceptor pair recombination emission (band B) appears. The peak energies of the band A and band B shift towards shorter wavelength side with increasing transport rate of (MeCp)$_2$Mg, corresponding to the increase in the band-gap energy due to the increase of the Mg composition. On the other hand, significant decrease of the intensity ratio of peak A to peak B is not observed. This means that the control of the Mg composition by (MeCp)$_2$Mg transport rate under suitable VI/II ratios, for example VI/II ratios of
one to two, has an advantage in achieving both the controllability of Mg composition and good optical property of undoped Zn$_{1-x}$Mg$_x$Te layer grown by atmospheric pressure MOVPE.

4. Conclusions
We have investigated the effects of the transport rates of DMZn and (MeCp)$_2$Mg upon the optical property of Zn$_{1-x}$Mg$_x$Te layer grown by atmospheric pressure MOVPE. The PL spectra exhibit two distinctive luminescence bands, i.e. excitonic emission and donor-acceptor pair recombination emission. The growth condition of suitable VI/II ratios is found to be essential for improving the PL property. The control of the Mg composition by (MeCp)$_2$Mg transport rate under high VI/II ratio of 1 and above has an advantage in achieving both the controllability of Mg composition and good optical property of undoped Zn$_{1-x}$Mg$_x$Te layer grown by atmospheric pressure MOVPE.

Acknowledgement
This study was partly supported by Industrial Technology Research Grant Program in 2005 from New Energy and Industrial Technology Development Organization (NEDO) of Japan.

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