ZnO is a wide-bandgap semiconductor with many attractive properties, such as a low-power threshold for optical pumping at room temperature and a large exciton binding energy of 60 meV, allowing highly efficient exciton recombination even at room temperature.\textsuperscript{15} ZnO and its ternary alloys have therefore attracted much attention due to their potential applications in optoelectronic devices operating in the blue and ultraviolet regions.\textsuperscript{14} An important step for the fabrication of ZnO-based LEDs is the realization of highly efficient quantum wells (QWs) and barrier layers. Structural and optical properties of ZnO/MgZnO multiple quantum wells (MQWs) grown on various substrates have been demonstrated by several groups.\textsuperscript{5–7} Most groups have reported on the ZnO/MgZnO MQWs grown by physical vapor deposition (PVD) methods such as molecular beam epitaxy (MBE) and pulsed laser deposition. However, there are no reports on the successful growth of ZnO/MgZnO MQWs by metallorganic chemical vapor deposition (MOCVD), except for rod-structured MQWs,\textsuperscript{9} due to the large lattice mismatch between MgO and ZnO, the difference in crystal structure of MgO and ZnO, and the low thermodynamic solubility limit of MgO in ZnO.\textsuperscript{10} MOCVD has well known advantages over physical processes, such as a good conformal coverage, epitaxial growth and selective deposition, the capability of large scale production, and growth of metastable materials.\textsuperscript{11} Gruber et al. investigated the properties of ZnO/MgZnO single QW grown by MOCVD and reported an increase in bandgap of 200 meV at a Mg concentration of 10% and an enhanced exciton binding energy of 96 meV.\textsuperscript{12} However, the detailed optical properties of ZnO/MgZnO MQWs grown by MOCVD are not yet fully understood. In this paper, we report on the successful growth of ZnO/MgZnO MQWs by MOCVD and the effect of quantum well thickness on the optical properties of MQWs.

**Experimental**

ZnO/MgZnO MQWs were grown on the sapphire substrate by using an MOCVD system with a vertical reactor. After the growth of a 60 nm-thick ZnO buffer layer on (0001) sapphire substrate by radio frequency (rf)-magnetron sputtering at 800 °C, a 400 nm undoped ZnO template was grown at 650 °C by MOCVD. Then, five periods of ZnO/MgZnO QWs were grown at 670 °C. Diethylylzinc (DEZn), bis(cyclopentadienyl)magnesium (Cp2Mg), and oxygen gas (99.9999% purity) were employed as the sources of zinc, magnesium, and oxygen, respectively. Ar gas (99.9999% purity) was used as a carrier gas. The metallorganic sources and O\textsubscript{2} were introduced into the reactor separately and mixed in the shower head, which was closely located 1 cm ahead of the substrate to minimize parasitic formation of ZnO particles in the gas phase. It was found that the MOCVD growth mode of ZnO film was changed from three-dimensional growth to two-dimensional growth at high VI/II gas ratio\textsuperscript{13} and a high VI/II gas ratio of 25,000 was employed in this study to enhance the lateral growth of the ZnO films. Three different ZnO/MgZnO MQWs with barrier thickness of 5 nm and well layer thicknesses (L\textsubscript{w}) of 0.7, 1.5, and 2 nm were grown to investigate the effect of quantum well thickness on the optical properties of MQWs. The quantum well thickness was determined from the PL spectra of MQWs with reported values of ZnO/MgZnO MQWs.\textsuperscript{14} The PL measurements were carried out using a He–Cd laser operating at a wavelength of 325 nm. To estimate the Mg composition of the barrier layers, MgZnO film was grown on an undoped ZnO template under the same growth conditions as the MQWs and the Mg composition was determined by PL measurement at 10 K. Based on the data given in Ref. 12, the Mg composition of the MgZnO barrier layers was estimated as 10%.

**Results and Discussion**

Scanning electron microscope (SEM) image of the ZnO/MgZnO MQWs showed a smooth and flat surface morphology, as shown in Fig. 1a. Figure 1b shows the cross-sectional TEM image of ZnO/MgZnO MQWs with L\textsubscript{w} of 2 nm. Modulation of the TEM image contrast clearly showed that the MQWs consisted of MgZnO and ZnO layers.

Figures 2a–2c show the temperature-dependent PL spectra of the ZnO/MgZnO MQWs with L\textsubscript{w} of 0.7, 1.5, and 2 nm. As shown in Fig. 2, the PL spectra of MQWs with L\textsubscript{w} of 0.7, 1.5, and 2 nm at 10 K show a MQW emission at 3.505, 3.424, and 3.406 eV, respectively. The energy of PL emission of MQWs with L\textsubscript{w} of 0.7, 1.5, and 2 nm is well agreed with the reported values of ZnO MQWs in the literature.\textsuperscript{14} The MQWs with L\textsubscript{w} of 0.7, 1.5, and 2 nm also show another peak at 3.356 eV at 10 K, which can be assigned to neutral donor-bound excitons from the undoped ZnO template layer,\textsuperscript{15} and the position of emission peak is not changed with L\textsubscript{w}. The PL peak of MgZnO barrier is also observed at 3.56 eV, showing that the Mg composition in the MgZnO barrier layer is 10%.\textsuperscript{12} The intensity of the quantum well emission compared to the donor-bound exciton is increased with increasing the...
Figure 2. Temperature dependence of PL spectra of ZnO/MgZnO MQWs with \( L_w \) of (a) 0.7 nm, (b) 1.5 nm, and (c) 2 nm.

\( L_w \). Moreover, the full width half maximum of MQWs emission at 300 K is decreased from 130 meV to 101 meV with increasing the \( L_w \) from 0.7 nm to 2 nm. The improved optical properties of MQWs are attributed to the improved uniformity of the quantum well thickness and alloy-potential of MQWs. The temperature-dependent changes of the PL peak energies of MQWs with \( L_w \) of 0.7, 1.5, and 2 nm are shown in Fig. 3. For the MQWs with \( L_w \) of 0.7 nm, the PL peak energy of the ZnO MQWs shows the S-shaped temperature dependence as the temperature increases from 10 to 300 K. The PL peak energy of MQWs with \( L_w \) of 0.7 nm red-shifts by 7 meV in the temperature range of 10–75 K. As the temperature further increases from 75 to 125 K, the peak energy blue-shifts by 19 meV. Above 125 K, the PL peak energy monotonically red-shifts due to a bandgap shrinkage. The S-shaped temperature dependence indicates that the ZnO quantum wells have the localized potential wells induced by the local variation in well-width or inhomogeneity of alloy-potentials in the MgZnO barrier layers.\(^8,15,16\) The S-shaped temperature dependence, however, gradually diminishes in the MQWs in the temperature range of 10–100 K with increasing the \( L_w \) from 1.5 nm to 2 nm, as shown in Fig. 3. These results indicate that the MQWs with \( L_w \) of 2 nm have much less localized potential wells in the ZnO well layer, which are induced by the local variation in well-width or inhomogeneity of alloy-potentials in the MgZnO barrier layers, compared to MQWs with \( L_w \) of 0.7 nm and 1.5 nm.

To investigate the internal quantum efficiency (IQE) of the ZnO/MgZnO MQWs, we measured the integrated PL intensities of the MQWs at temperatures of 10–300 K as shown in Fig. 4. The IQEs were then estimated by comparing the temperature-dependent integrated PL intensities, assuming that IQE is 100% at the low temperature of 10 K regardless of excitation carrier density.\(^17\) The IQE of ZnO/MgZnO MQWs with \( L_w \) of 0.7, 1.5, and 2 nm at 300 K was estimated to be 2.8%, 4.1%, and 4.9%, respectively. This result indicates that the defect density of the MQWs is decreased with increasing the \( L_w \) of MQWs. It is noted that the IQE of ZnO/MgZnO MQWs is increased with decreasing the S-shaped PL property. In the case of InGaN/GaN MQWs, the S-shaped PL property is caused by the fluctuation of In composition in the quantum well layer, and this inhomogeneity of composition in the quantum well enhances the IQE by capturing the electrons and holes in the localized potential wells before carriers are captured by the nonradiative recombination centers.\(^18,19\) In the case of ZnO/MgZnO MQWs, however, the IQE was improved with decreasing the S-shaped property. These results indicate that the low defect density and high uniformity and homogeneity of MQWs are essential factors to improve the IQE of ZnO/MgZnO MQWs because the S-shaped PL property is resulted from the variation in well thickness or inhomogeneity of alloy-potentials in the MgZnO barrier layers. The S-shaped property can be reduced by improving the uniformity of the quantum well thickness and alloy-potential of MQWs. Two-step growth, growth interruptions, and repeated temperature modulation
processes\textsuperscript{20–22} are expected to further improve the uniformity and homogeneity of MQWs.

Conclusions

In summary, we have investigated the optical properties of ZnO/MgZnO MQWs grown by MOCVD. The temperature-dependent PL spectra of MQWs showed that the S-shaped PL property is reduced with increasing the well thickness from 0.7 to 2 nm. The IQE of the MQWs with $L_w$ of 2 nm was higher by 75\% than that of the MQWs with $L_w$ of 0.7 nm. The high IQE of ZnO/MgZnO MQWs with $L_w$ of 2 nm is attributed to a low defect density and more uniform layer thickness or alloy composition of MgZnO barrier layer, compared to MQWs with $L_w$ of 0.7 and 1.5 nm.

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