Extended Abstracts of the 2002 International Conference on Solid State Devices and Materials, Nagoya, 2002, pp. 528–529

P5-14

Epitaxial growth of ZnO films by low-pressure MOVPE

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Realization of single crystalline ZnO films is of great importance to ultra-violet laser emitting devices. In addition, due to the small lattice mismatch to GaN, ZnO films can be used as good substrates for GaN epitaxial growth. ZnO epitaxial films have been grown by molecular beam epitaxy (MEB), metal organic vapor phase epitaxy (MOVPE), and sputtering. Among various techniques, MOVPE has advantages of both large-area deposition and high quality which are essential to industrial applications. Up to now, however, there are only a few reports on epitaxial growth of ZnO films by MOVPE.[1-4]

Here, we report the epitaxial growth of ZnO films by MOVPE on sapphire (0001) substrates at 6 Torr and 0.05 Torr. Precursors were diethyl zinc (DEZn) and oxygen gas. The carrier gas for Zn was high-purity nitrogen gas. The precursors are different from those in Refs.[1,2] while the carrier gas is different from that in Refs.[3,4]. The flow of O_2 was set to be 20 ~ 40 sccm and that of nitrogen was 2.5 ~ 10 sccm. The temperature of DEZn was 7 ~ 15 °C. The substrate temperature was varied from 400 to 500 °C. Photoluminescence (PL) measurements were carried out under the excitation of a He-Cd laser (325 nm) at temperatures of 4 ~ 300 K. Reflection spectra were obtained using a xenon lamp at normal incidence. Transmission spectrum was obtained using a photo spectrometer at room temperature.

Characterization using x-ray diffraction demonstrated that the films were grown with c-orientation, i.e., ZnO(0001)//Al_2O_3(0001). The best value of full width at half maximum (FWHM) of the rocking curve of the ZnO (0002) plane was 0.2°, being comparable to those of other MOCVD method on (0001) sapphire and of molecular beam epitaxy (MBE) method on (0001) GaN. The ϕ scan of ZnO $(11\bar{2}2)$ planes obtained by rotating the sample around its surface normal was featured with six diffraction peaks, demonstrating that the films were grown epitaxially. The epitaxial relationship between ZnO films and sapphire substrates was investigated. We found that the in-plane orientation of the ZnO films can be easily controlled by selecting the initial growth layer, i.e., ZnO $[10\bar{1}0]$ // sapphire $[11\bar{2}0]$ when the growth is oxygen-started and ZnO $[10\bar{1}0]$ // sapphire $[10\bar{1}0]$ when Zn-started. This supposes the possibility of controlling the surface polarity of the ZnO films by MOVPE.

Distinct emissions from free *A*-exciton in these ZnO epitaxial films were observed at 3.5 K with a line width as small as 3 meV. With rise in temperature, the emission intensity of *A* excitons increased relative to that of bound excitons and dominated the emission from 60 K to room temperature. Transitions associated with *A* and *B* excitons were also observed clearly in reflection spectra at low temperatures. Absorption due to free excitons was observed at room temperature. These results demonstrate the good quality of the film.

[1] K. Ogata et al, phys. stat. sol. (b) 229, 915(2002). [2] Y. Kashiwaba et al, phys. stat. sol. (b) 229, 921(2002).

[3] C. R. Gorla et al, J. Appl. Phys. 85, 2595(1998). [4] W. I. Park et al, Appl. Phys. Lett. 79, 2022(2001).

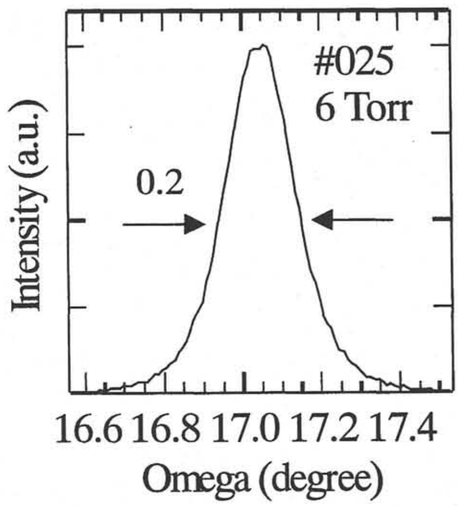

Fig. 1 X-ray rocking curve of (0002) plane of a ZnO film grown at 6 Torr. FWHM=0.2 degree.

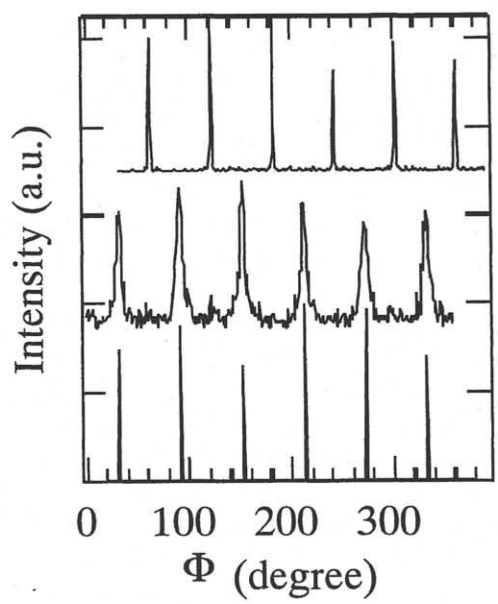

Fig. 2 Φ scan of ZnO (112) and sapphire (116) planes.
Top: ZnO grown by O-started mode.
Middle: ZnO grown by Zn-started mode.
Bottom: sapphire substrate.

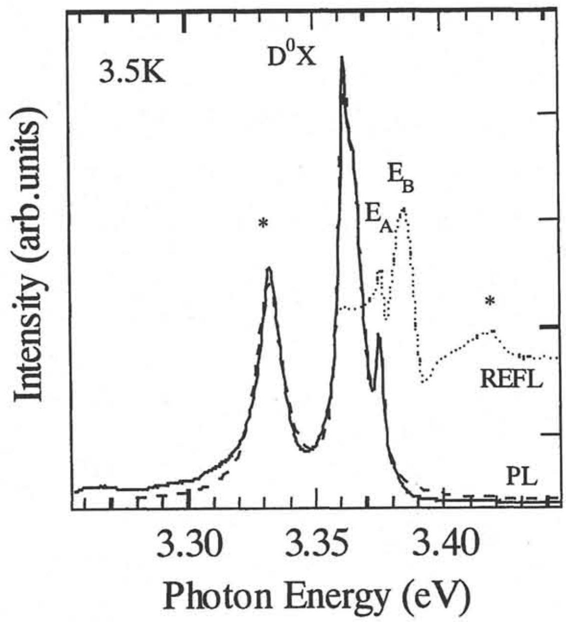

Fig. 3 Low-temperature PL and reflection spectra. Dashed curve is the fitted result of the PL spectrum. Free excitonic transitions were observed from both PL and reflection spectra.

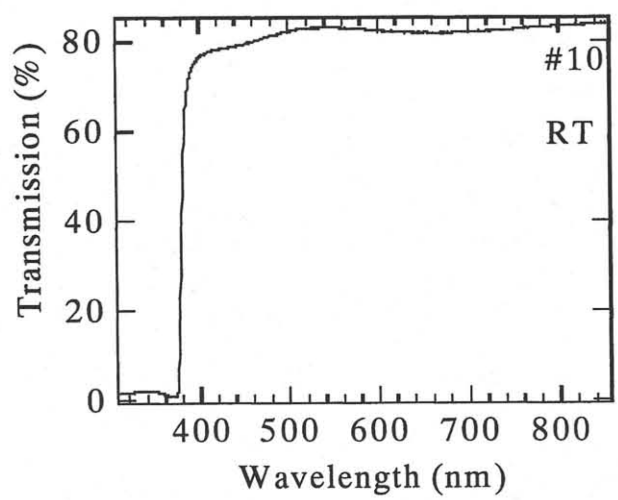

Fig. 4 Typical transmission spectrum of the ZnO epitaxial films.