Investigation of MgZnO/ZnO Heterostructures Grown on c-Sapphire Substrates by Pulsed Laser Deposition

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ABSTRACT

MgZnO thin films were grown on c-sapphire and ZnO-coated c-sapphire substrates by pulsed laser deposition from a ZnMgO target with 4 at% Mg. The MgZnO grown on the ZnO underlayer showed significantly better crystal quality than that grown directly on sapphire. AFM studies revealed a significant deterioration in surface morphology for the MgZnO layers compared with the ZnO underlayer. Optical transmission studies indicated a MgZnO bandgap of 3.61eV (compared with 3.34eV for the ZnO), which corresponds to a Mg content of about 16.1 at%. The MgZnO/ZnO heterojunction showed an anomalously low resistivity, which was more than two orders of magnitude less than the MgZnO layer and an order of magnitude lower than that for the ZnO layer. It was suggested that this may be attributable to the presence of a 2D electron gas at the ZnMgO/ZnO heterointerface.

Keywords: ZnO, MgZnO, heterointerface, PLD, UV, Transmittance, bandgap engineering

1. INTRODUCTION

ZnO has been investigated extensively in recent years for use in a huge range of potential applications based on its' distinctive and tuneable property set [1,2]. One of the most interesting aspects of this is the combination of a direct wide bandgap (Eg ~ 3.4 eV) with a relatively high exciton binding energy (60meV) [3]. This opens up the perspective of devices such as solar-blind ultraviolet (UV) photodetectors [4] and efficient UV emitters with low lasing thresholds [5,6]. In this paper, we investigate the alloying of ZnO with MgO (Eg ~ 7.7 eV) in order to extend the ZnO bandgap further into the UV [7,8]. Such alloying has generated significant interest of late for use in MgZnO/ZnO/MgZnO quantum wells [9] and because of the creation of two dimensional electron gases (2DEG) at ZnO/MgZnO heterointerfaces due to a strong built-in potential arising from polarization mismatch [10]. Such 2DEG samples show remarkably high mobilities (in excess of 300 000 cm^2/Vs at 40mK [11]) and exhibit the fractional quantum Hall effect [12,13]. In this study we investigate the impact of Mg doping on ZnO films and use this as a basis for interpreting the structural, optical and electrical response of the MgZnO/ZnO heterointerface.

2. EXPERIMENT

MgZnO thin films were grown simultaneously on *c*-Al_2O_3 and ZnO-coated *c*-sapphire substrates using pulsed laser deposition (PLD). The PLD growth employed commercial targets (sintered ZnO and MgZnO (4 at% Mg)) and was conducted in molecular oxygen with a coherent KrF (λ = 248 nm) excimer laser, using conditions described elsewhere [14]. The crystal structure of the samples was investigated using high resolution X-Ray Diffraction (XRD) performed in a Panalytical MRD Pro system using Cu $K\alpha_1$ radiation. Surface morphology was investigated using Veeco Innova Atomic Force Microscope (AFM) in tapping mode. Scanning Electron Microscopy (SEM) was conducted in an FEI field emission gun XL30S system. Optical absorption was investigated using a Perkin-Elmer Lambda 950 UV/VIS/NIR spectrometer system. Resistivity measurements were made using co-linear 4 point probe current/voltage measurements (for both current directions) at several areas on each sample

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3. RESULTS & DISCUSSION

Figure 1 shows XRD 2 theta-omega scans for the (0002) peak of the ZnO/sapphire, MgZnO/sapphire and MgZnO/ZnO/sapphire samples.

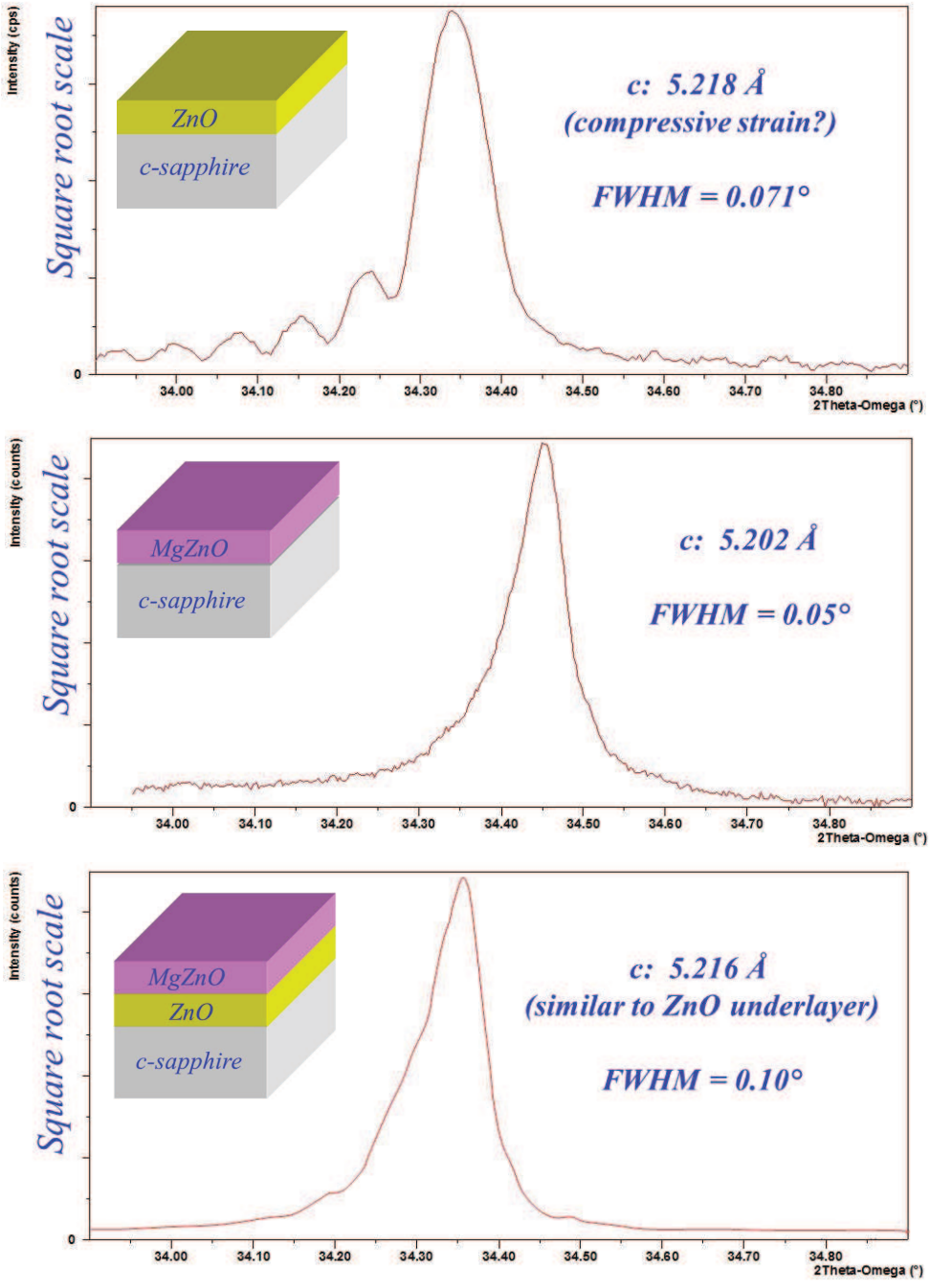

Figure 1. XRD 2 theta-omega scans of the (0002) peak for ZnO/sapphire, MgZnO/sapphire and MgZnO/ZnO/sapphire.

For the ZnO/sapphire, the 2 theta-omega peak position corresponds to a c-lattice parameter of 5.218 Å (as compared to an equilibrium value of about 5.206 Å). This suggests that the film was under compressive strain in the a-b plane. The scan also shows Pendellusong fringes, which indicate that the surface was relatively smooth over the scale of the diffraction spot (few mm^2). The fringe spacing gives an estimate of film thickness at about 110 nm. The MgZnO/sapphire peak position indicates a c-lattice parameter of 5.202 Å. This is close to the ZnO equilibrium value, which is consistent since the substitutional Mg^{2+} ion has a fairly similar ionic radius to the Zn^{2+} ion [15]. There are no Pendellusong fringes in this case, which implies that the MgZnO surface morphology is rougher than that for the ZnO layer. The MgZnO/ZnO/sapphire peak position corresponds to a c-lattice parameter of 5.216 Å, which is significantly larger than the value found for MgZnO/sapphire and similar to that for the ZnO/sapphire. This suggests that there has been epitaxy on the ZnO underlayer and that the MgZnO overlayer is also strained. Once again, there are no Pendellusong fringes, thus there appears to be a degradation in surface morphology compared to the underlying ZnO surface (although the sample is thicker, so some suppression of the fringes can be expected independent of surface quality).
The XRD omega-scan rocking curve linewidths (high resolution optics) are 0.006º, 0.15º and 0.06º, respectively, for the ZnO, MgZnO and MgZnO/ZnO. This indicates that the dispersion in the crystallographic orientation about the c-axis was higher for the MgZnO samples compared to the ZnO. The MgZnO layer grown on the ZnO underlayer, however, was significantly more c-axis oriented than the MgZnO layer grown directly on sapphire. Overall, the XRD results indicate that incorporation of Mg generally degrades the crystal quality compared to ZnO itself (as reported elsewhere [16]) but that use of a ZnO underlayer considerably improves the MgZnO layer quality.

Figure 2 shows an SEM image of a fracture cross section of the MgZnO/sapphire sample. This gives an estimate of film thickness at about 260 nm.

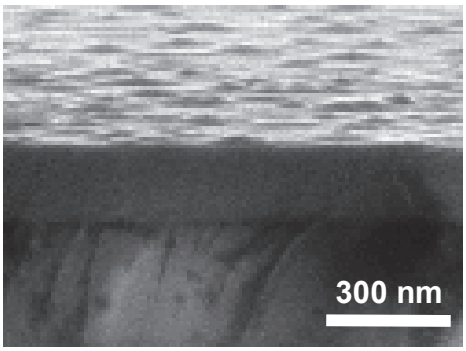

Figure 2. SEM image of a fracture cross-section of the MgZnO/sapphire sample.

Figure 3 shows tapping mode AFM images for all three samples. The ZnO layer has the lowest surface roughness (in agreement with the XRD findings) with a Root Mean Square (RMS) roughness of 0.9 nm (over an area of ~1μm x 1μm). The MgZnO and MgZnO/ZnO layers have rougher surfaces with RMS values of 8.5 and 5.5 nm, respectively. These RMS values integrate comparatively large peak-to-peak figures due to the presence of asperities on a smoother underlying surface, so it is difficult to differentiate between the MgZnO and MgZnO/ZnO based on these figures.

Figure 4 shows optical transmission spectra for the ZnO, MgZnO/sapphire and MgZnO/ZnO/sapphire samples. The spectra all display excellent transparency in the visible (N.B. the spectra integrate supplementary losses due to the sapphire substrate) as expected for a material that is often adopted for transparent electrode applications [17]. All samples also show a UV cut-off. The principal cut-off wavelength for the ZnO/sapphire and MgZnO/sapphire samples is similar and corresponds to an energy of ~3.34eV. This is as would be expected for the bandgap of the ZnO layer, which is present in both samples. The MgZnO/sapphire sample shows a UV cut-off which corresponds to an energy of ~3.61eV. This is consistent with an Mg content of 16.1 $\pm$ 0.4 at% [18,19], which is 12.1 at% more than the nominal value in the MgZnO PLD target and significantly in excess of the 4 mol% solubility limit for Mg based on the equilibrium MgO/ZnO binary system phase diagram [20]. Such an Mg enhancement compared to the target concentration has been reported elsewhere for such non-equilibrium PLD growth of MgZnO and was attributed to a difference in vapor pressures [16]. Another interesting feature of the transmission curves is that the ZnO layer does not give complete extinction while the MgZnO layer does. This is most probably due to the higher absorbing thickness (~260 nm) of the MgZnO layer compared to that of the ZnO layer (~110 nm).

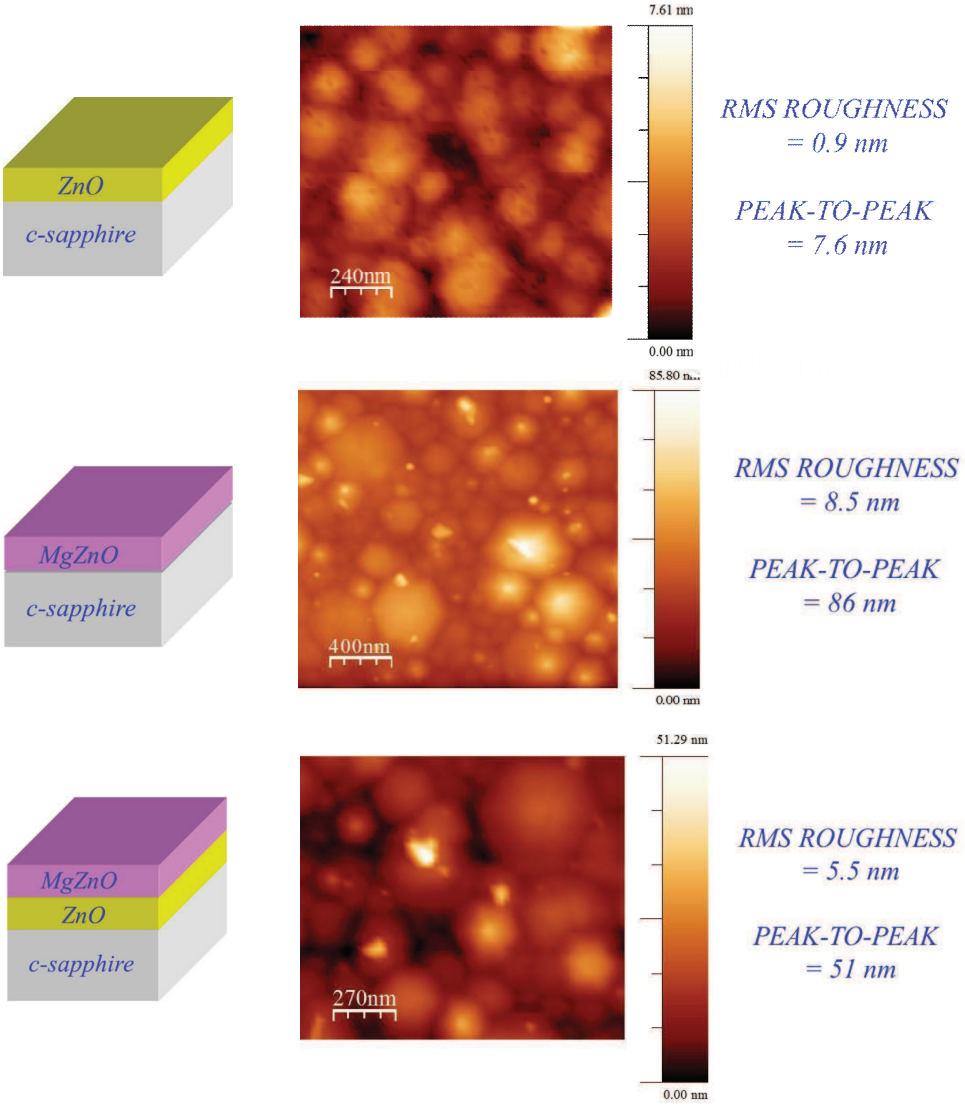

Figure 3. Tapping mode AFM images of the ZnO/sapphire, MgZnO/sapphire and MgZnO/ZnO/Sapphire surface morphologies.

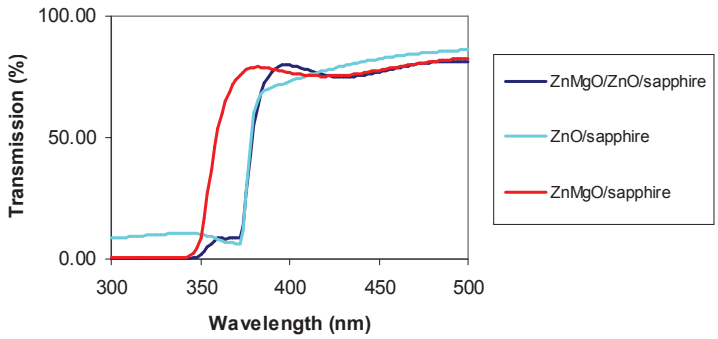

Figure 4. Optical transmission spectra for the ZnO, MgZnO/sapphire and MgZnO/ZnO/sapphire samples.

Table 1 shows the resistivity values measured for each sample.

	ZnO/c-sapphire	*MgZnO/c-sapphire*	*MgZnO/ZnO/c-sapphire*
Resistivity (Ω.cm)	0.05 ± 0.001	0.85 + 0.01	0.006 + 0.002

Table 1. Resistivity values for the ZnO/sapphire, MgZnO/sapphire and MgZnO/ZnO/sapphire samples.

The resistivity for the MgZnO/sapphire is higher than for ZnO/sapphire grown under similar conditions, as reported elsewhere for PLD growth [16]. The resistivity for the MgZnO/ZnO/sapphire layer, however, is more than two orders of magnitude lower than that for the MgZnO and an order of magnitude lower than that for the ZnO/sapphire. This resistivity value, of 0.006 ohm.com, is exceptionally low in absolute terms for a ZnO or MgZnO sample without intentional shallow donor doping [16]. The reproducibility of the value was confirmed by measuring several areas of sample at various forward and reverse injection currents. A similar drop in RT resistivity was reported for ZnMgO/ZnO heterostructures elsewhere and found to be due to the presence of a 2DEG. Temperature dependent Hall measurements will be conducted to further explore this [21].

4. CONCLUSIONS

MgZnO thin films were grown on c-sapphire and ZnO-coated c-sapphire substrates by PLD. XRD revealed that the MgZnO grown on the ZnO underlayer had significantly better crystal quality than that grown directly on sapphire. AFM studies revealed a significant deterioration in surface morphology for the MgZnO layers compared with the ZnO underlayer. Optical transmission studies indicated a MgZnO bandgap of 3.61eV (compared with 3.34eV for the ZnO), which corresponds to a Mg content of about 16.1 at%. The MgZnO/ZnO/sapphire sample showed a resistivity of 0.006 Ω.cm, which was more than two orders of magnitude lower than for the MgZnO layer and an order of magnitude lower than that for the ZnO layer. It was suggested that this enhanced conductivity might be attributable to the presence of a 2DEG at the ZnMgO/ZnO heterointerface.

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