Characterization of ZnO Thin Films Grown on c-Sapphire by Pulsed Laser Deposition as Templates for Regrowth of ZnO by Metal Organic Chemical Vapor Deposition

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ABSTRACT

The use of ZnO template layers grown Pulsed Laser Deposition (PLD) has been seen to produce dramatic improvements in the surface morphology, crystallographic quality and optical properties of ZnO layers grown on c-sapphire substrates by Metal Organic Chemical Vapor Deposition. This paper provides complementary details on the PLD-grown ZnO template properties.

Keywords: ZnO, PLD, Template, MOCVD, Regrowth

1. INTRODUCTION

ZnO thin films are currently of strong research interest for a very wide range of existing and emerging applications[1]. Many authors have adopted Metal Organic Chemical Vapor Deposition (MOCVD) for these studies because of its' proven industrial track record for reliable, high throughput, production of opto-electronic-grade compound semiconductor materials with complex alloying and doping profiles. The majority of the earlier MOCVD studies focused on the growth of ZnO layers on c-sapphire (c-Al_2O_3) substrates because of their compatibility, high quality, availability, low cost and wide areas. Various authors reported, however, that it was difficult to obtain good quality ZnO layers directly on c-Al_2O_3 by MOCVD[2].

Recently, there have been significant breakthroughs in the quality of commercially-available bulk ZnO substrates[3]. These have given much better results for MOCVD growth of ZnO than c-Al_2O_3[4]. Unfortunately, the availability of high quality 2 inch diameter ZnO substrates remains very poor and the cost level is prohibitive for most practical applications. Moreover, the perspectives for the commercialization of larger-area bulk ZnO substrates are unclear for the moment.

Pulsed Laser Deposition (PLD) has been widely adopted for the growth of ZnO, and oxide materials in general[5]. In contrast to MOCVD, PLD has demonstrated a capacity to grow excellent ZnO layers directly on c-Al_2O_3. Recently, the authors reported that superior quality ZnO layers could be obtained on c-Al_2O_3 substrates by low temperature MOCVD (420°C), through the use of ZnO template layers grown by PLD[6] (Figure 1). It was found that the quality of the obtained MOCVD layers was even equivalent to that for growth on commercial hydrothermal ZnO substrates in terms of the surface morphology, crystallographic quality and optical properties.

This paper gives supplementary characterization data on the PLD-grown ZnO templates, which were used for the MOCVD regrowth.

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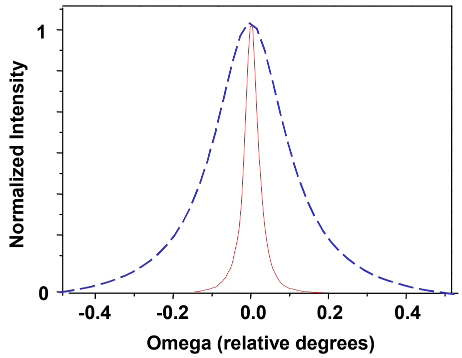

Figure 1 XRD Ω rocking curves for the (0002) peaks of 2.5μm thick ZnO films grown on c-Al_2O_3 by MOCVD with (solid) and without (dashed) a PLD-grown ZnO template layer[6]. Full Wave Half Maxima (FWHM) are 119 and 828 arc.sec, respectively. This shows that the film grown on the PLD template had much less dispersion in the orientation about the crystallographic c-axis.

2. EXPERIMENT

The ZnO template layers were grown on c-Al_2O_3 substrates by ablation of a 5N ZnO target with a KrF excimer laser (248nm), as described elsewhere[7]. X-ray diffraction (XRD) studies were conducted with a 4-circle Philips X-Pert MRD PRO system. Atomic Force Microscopy (AFM) was conducted using a Digital Instruments D3100 system. MCs+ Secondary Ion Mass Spectroscopy (SIMS) was conducted using a Cameca IMS4F system. The primary (incident) ions were 133Cs+, with an impact-energy of 5.5 keV. Positively charged secondary ions were detected.

3. RESULTS

After optimizing each growth parameter, ZnO thin film templates of high crystallographic, morphological and optical quality were obtained. Figure 2 shows the mirror-like nature of the film surface.

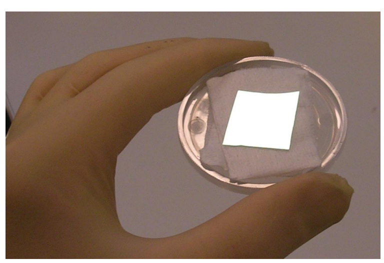

Figure 2: Photograph showing the mirror-like surface of the ZnO film grown by PLD on c-Al_2O_3.

Figure 2 shows a 2θ/Ω scan for the (0002) peak of the ZnO grown on c-Al_2O_3 by PLD. The very strong peak suggests that the film is highly c-axis oriented. The peak has a FWHM of 120 arc.sec and a maximum corresponding to a c lattice parameter of 5.213Å, which is a little larger than would be expected for relaxed wurtzite ZnO. This implies that the layer might be compressively strained in the film plane. The scan also shows extensive Pendellosung fringes, indicative of a smooth surface morphology. The fringe spacing gives an estimate of film thickness at about 300 nm

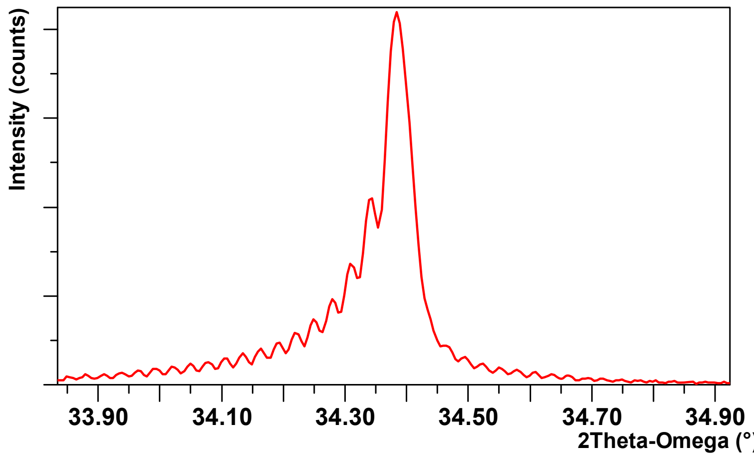

Figure 3: X-ray diffraction 2θ/Ω scan for the (0002) peak of the ZnO film grown by PLD on c-Al_2O_3. (intensity in square root scale).

The (0002) peak Ω scans (Figure 4) show FWHM of 29 and 140 arc.sec in "high resolution" and "open detector" modes, respectively. These values indicate that the film has a very small dispersion in the crystallographic orientation about the c axis.

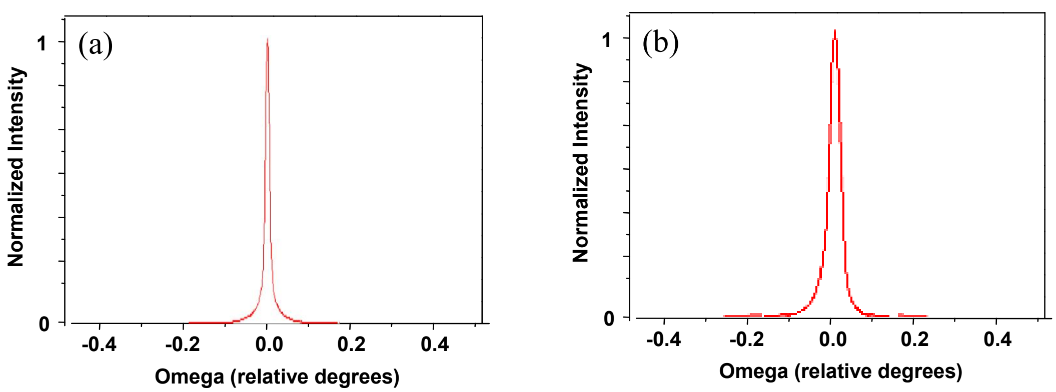

Figure 4: XRD Ω scans for the (0002) peak of the ZnO film grown by PLD on c-Al_2O_3 with (a) a 3 bounce Ge monochromator and (b) open detector.

Figure 5: shows an AFM image of a typical area of the ZnO film grown on c-Al_2O_3 by PLD. Root mean square roughness over an area of 2.5µm x 2.5µm was < 1 nm.

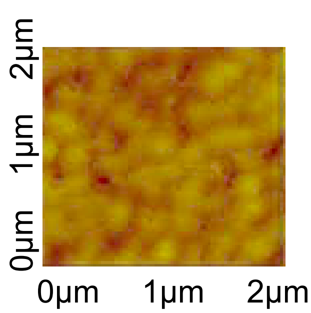

Figure 5: AFM image of ZnO film grown by PLD on c-Al_2O_3.

Figure 6 shows Al concentration profiles along the growth direction for ZnO grown directly on c-Al_2O_3 by PLD and MOCVD (Al is a shallow donor which can compensate attempts to p-dope ZnO). The Al concentration for the film grown by PLD drops off much more abruptly in the ZnO layer (by 4 orders of magnitude within 50nm from the ZnO/c-Al_2O_3 interface, compared with over 200nm for an equivalent drop off in the MOCVD-grown layer). This suggests that the PLD grown layer is less susceptible to interdiffusion of Al from the c-Al_2O_3 substrate. It has been proposed that this may be due to a more dense (less polycrystalline) structure providing a better diffusion barrier.

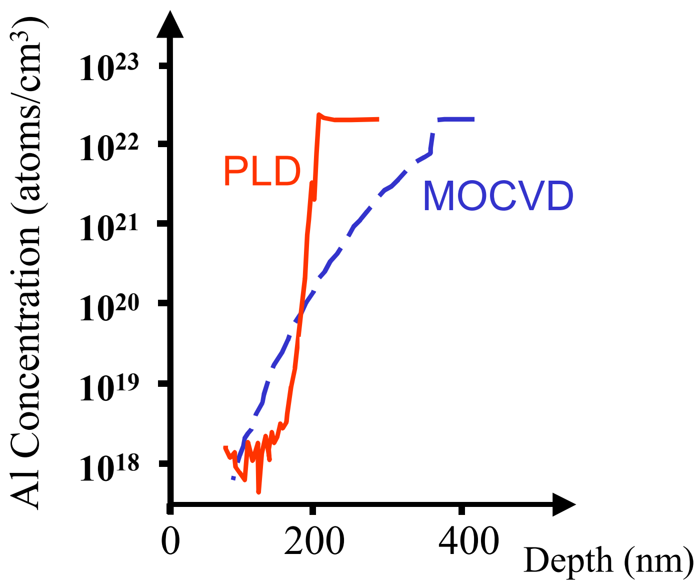

Figure 6: SIMS depth profiles for Al along the growth direction in ZnO deposited directly on c-Al_2O_3 by PLD and MOCVD (N.B. film thicknesses are different).

5. CONCLUSION

300 nm thick ZnO films were grown on c-Al_2O_3 substrates by PLD. XRD and AFM studies indicate that these layers have very high crystallographic quality and smooth surface morphology. SIMS profiling suggests that they may also provide a barrier to Al diffusion from the c-Al_2O_3 substrate. The use of such films as templates for MOCVD ZnO regrowth produced dramatic improvements in crystallographic, morphological and optical quality compared with growths directly on c-Al_2O_3 substrates[6]. Thus PLD-grown templates could be a promising low-cost and large area alternative to bulk ZnO substrates for MOCVD growth of ZnO.

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REFERENCES

1. C. Klingshirn, Chem. Phys. Chem. 8 (2007) 782 – 803
2. V. Sallet, C. Thiandoume, J.F. Rommeluere, A. Lusson, A. Riviere, J.P. Riviere, O. Gorochov, R. Triboulet, V. Munoz-Sanjose, Materials Letters 53 (2002) 126–131

3. K. Maeda, M. Sato, I. Niikura & T. Fukuda, Semicond. Sci. Technol. 20 (2005) S49–S54
4. C. Neumann, S. Lautenschläger, S. Graubner, J. Sann, N. Volbers, B. K. Meyer, J. Bläsing, A. Krost, F. Bertram, J. Christen, Phys. Stat. Sol. 244, 5 (2006) 1451 - 1457
5. M. Lorenz *Pulsed Laser Deposition of ZnO-based Thin Films* chapter 7 in: K. Ellmer, A. Klein, B. Rech (eds.), "Transparent Conductive Zinc Oxide. Basics and Applications in Thin Film Solar Cells" (Springer Series in Materials Science Vol. 104, Berlin, 2007) 305-360
6. C. Sartel, V. Sallet, A. Lusson, N. Haneche, J. M. Laroche, P. Galtier, D. J. Rogers, F. Hosseini Teherani, submitted to J. Vac. Sci. Technol. (2009)
7. D. J. Rogers, F. Hosseini Téherani, A. Yasan, R. McClintock, K. Mayes, S. R. Darvish, P. Kung, M. Razeghi, and G. Garry, Proc. SPIE 5732 (2005) 412

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