ELSEVIER

Thin Solid Films 259 (1995) 1–4

thin solid films

Letter

Growth of ZnO thin films on GaAs by pulsed laser deposition

V. Craciun[a], J. Elders[b], J. G. E. Gardeniers[b], J. Geretovsky[a], Ian W. Boyd[a]

[a]*Electronic and Electrical Engineering, University College London, London WC1E 7JE, UK*
[b]*MESA Research Institute, University of Twente, PO Box 217, 7500 AE Enschede, Netherlands*

Received 13 September 1994; accepted 21 December 1994

Abstract

ZnO thin films have been grown on GaAs substrates using the pulsed laser deposition technique with or without a photodeposited SiO_2 buffer layer. The presence of the SiO_2 layer has a beneficial effect on the crystalline quality of the grown ZnO films. Highly *c*-axis oriented ZnO films having a full width at half maximum value of the (002) X-ray diffraction line of less than 0.13° have been grown on such buffer layers at a substrate temperature of only 350 °C.

Keywords: Laser ablation, Zinc oxide; X-ray diffraction; Deposition process; Structural properties

1. Introduction

ZnO is a well studied n-type semiconductor which crystallises in the wurtzite lattice ($c = 0.521$ nm, $c/a = 1.60$ [1]). It is optically transparent in the visible region of the spectrum ($E_g = 3.26$ eV), its electrical conductivity can be readily modified by orders of magnitude by doping with various group III and group IV elements and, when *c*-axis oriented, it exhibits good piezoelectric and piezo-optic properties and a large piezoelectric coupling coefficient [1, 2]. These attractive properties have been exploited in numerous applications such as gas sensors, solar cells, optically active devices, actuators, varistors, etc. [3–7].

Irrespective of the deposition technique, there is a general consensus for the need of minimising substrate temperatures without sacrificing crystalline quality [8, 9]. This requirement is especially stringent for opto-acoustic device applications, where the substrate choice of GaAs [9–12] and a processing temperature below 450 °C are essential.

It has already been shown that the pulsed laser deposition (PLD) method can be used to grow high-quality *c*-axis oriented ZnO films on Si or glass substrates at lower temperatures than most other techniques [13, 14]. In this letter we show that by employing the optimised deposition conditions previously determined [14], ZnO films having a crystalline structure amongst the best reported so far by any technique can also be grown on GaAs substrates using the PLD method.

2. Experimental details

Cylindrical ZnO targets were ablated by 20 ns pulses from a KrF laser ($\lambda = 248$ nm) within our PLD set-up, which has been described in detail elsewhere [13, 14]. The deposition conditions were similar to those which were found to produce the best quality ZnO films on glass or Si substrates: laser fluence, 2.1 J cm^{-2}; oxygen (99.999% purity) working pressure, 1.33×10^{-3} Torr; and substrate temperature, 300–350 °C. The films were deposited on GaAs substrates placed on a heater situated 4 cm in front of the target. As it has been previously shown for sputtered ZnO films that a thin buffer layer of SiO_2 can alleviate stress problems and promote a good texture [10–12], some of the GaAs substrates were coated with a thin SiO_2 layer before ZnO deposition. The silica films were photodeposited from a SiH_4

SSDI 0040-6090(94)09479-9

(0.50 sccm) and N_2O (50.0 sccm) mixture under the irradiation of a Xe excimer lamp ($\lambda = 174$ nm) at a substrate temperature of 250 °C [15, 16]. The total pressure during the deposition was 10 mbar and the thickness of the deposited SiO_2 layers, measured by ellipsometry, was 12 ± 1 nm.

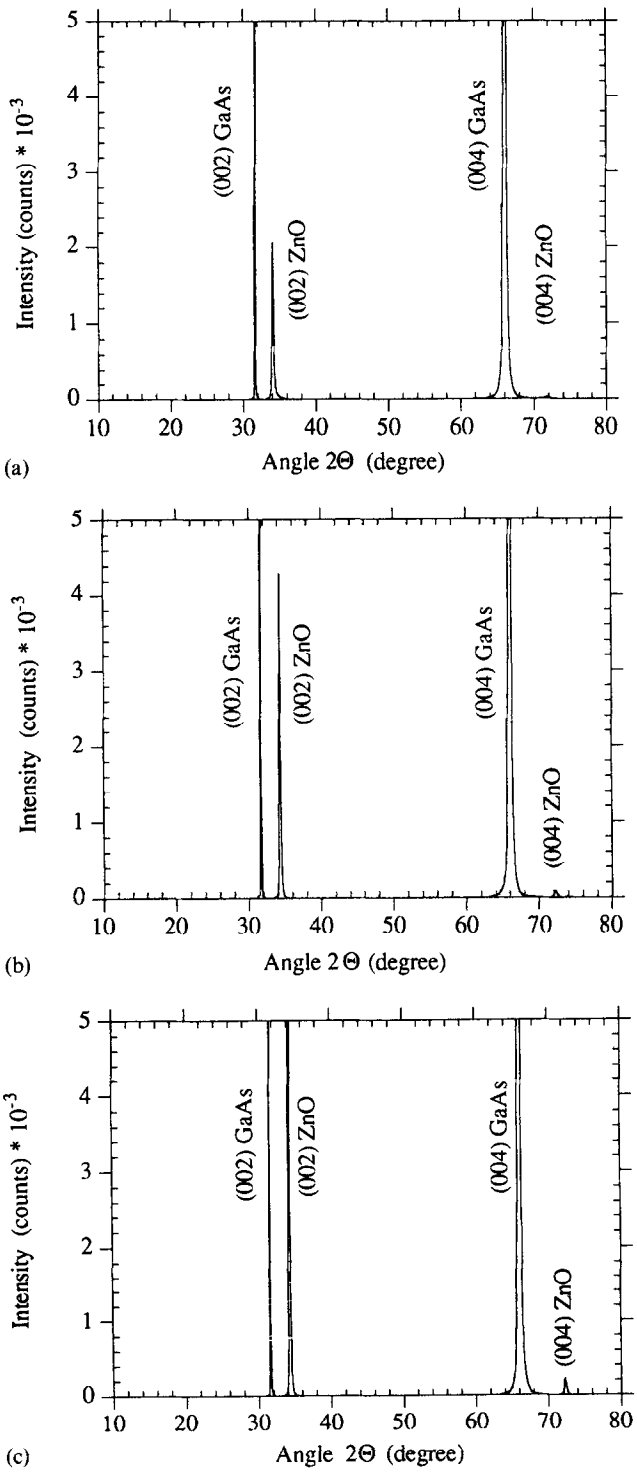

Fig. 1. XRD patterns of a ZnO thin film deposited on GaAs at 300 °C (a), 350 °C (b) and on SiO_2/GaAs at 350 °C.

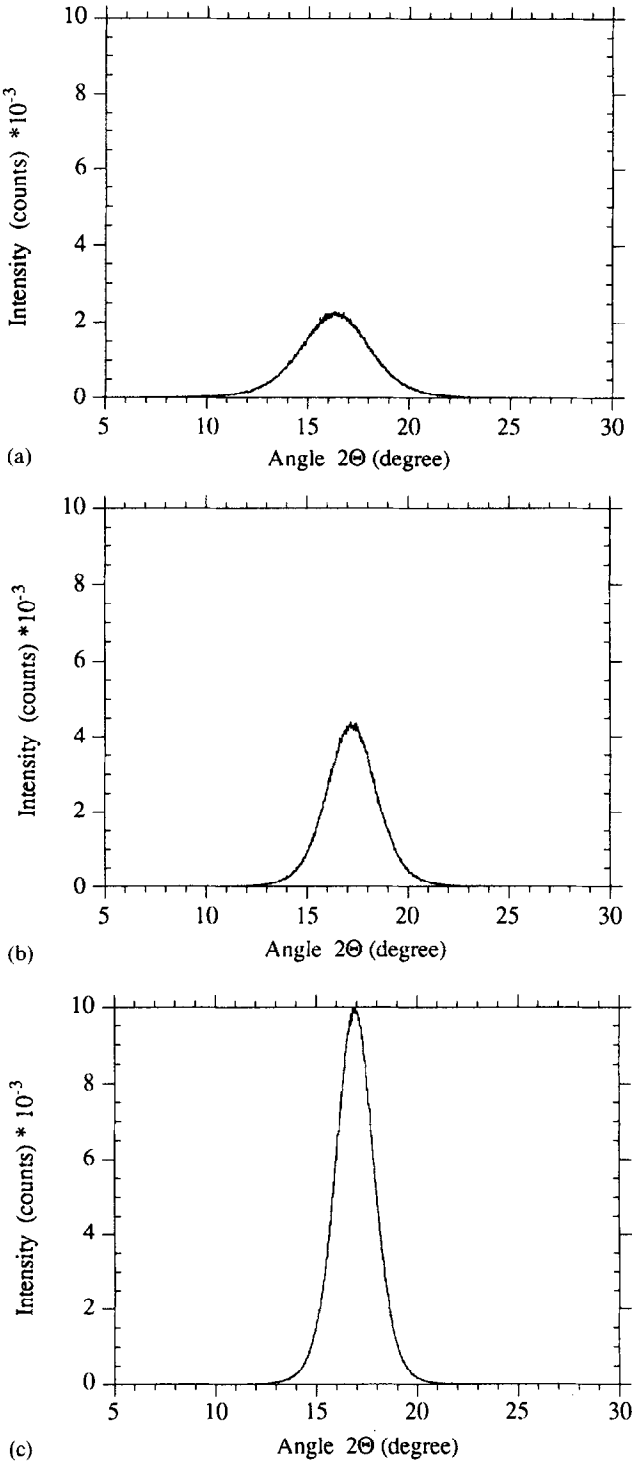

Fig. 2. Rocking curves of the ZnO films shown in Fig. 1.

The crystalline structure of the grown ZnO films was investigated by X-ray diffraction (XRD) and an X-ray rocking curve. The Cu Kα source used emitted three lines: K$\alpha_1 = 0.154060$ nm and K$\alpha_2 = 0.154439$ nm (intensity half that of Kα), as well as K$\beta = 0.139222$ nm, which was removed by placing a filter in the X-ray beam. The surface morphology and cross-sectional

Table 1
XRD and X-ray rocking curve results for ZnO films deposited under various conditions

Sample	T_s (°C)	(002) line		$I_{(002)}$ZnO/$I_{(002)}$GaAs	Rocking curve FWHM (002) (deg)	c_{axis} (nm)	Strain (⊥surface) (%)
		2Θ (deg)	FWHM (deg)				
ZnO/GaAs	300	34.09	0.250	0.17	3.965	0.5260	1.03
ZnO/GaAs	350	34.31	0.202	0.37	2.854	0.5228	0.41
ZnO/SiO_2/GaAs	350	34.36	0.176	0.78	2.187	0.5220	0.26

structure of fractured films was investigated using scanning electron microscopy (SEM).

In Fig. 1, the XRD spectra recorded for ZnO films grown at substrate temperatures of 300 and 350 °C on GaAs substrates with and without the buffer SiO_2 layer are shown. The films are highly textured, only the ZnO (002) and (004) XRD lines being present along with the GaAs substrate lines. It should be mentioned that non-c-axis orientation peaks are much stronger than the (002) peak in XRD data for ZnO powder [17]. The position, width and intensity of the XRD lines and the corresponding c-axis lattice values and macro-strains are presented in Table 1. From the results shown in Table 1 one can see that the increase of the substrate temperature from 300 to 350 °C has a beneficial effect both on crystallinity and stress. Even further improvement of the crystalline quality is obtained by the presence of the buffer SiO_2 layer, as is found with similar reports for sputtered ZnO films [10–12]. The full width at half maximum (FWHM) of the (002) reflection line, corrected for the instrumental broadening as derived from the width of the GaAs (002) peak for the ZnO/SiO_2/GaAs sample grown at 350 °C, is only 0.13°, one of the lowest values yet reported in the literature [2–13]. The average crystalline size corresponding to this linewidth estimated from the Scherrer equation [18] is well above 50 nm.

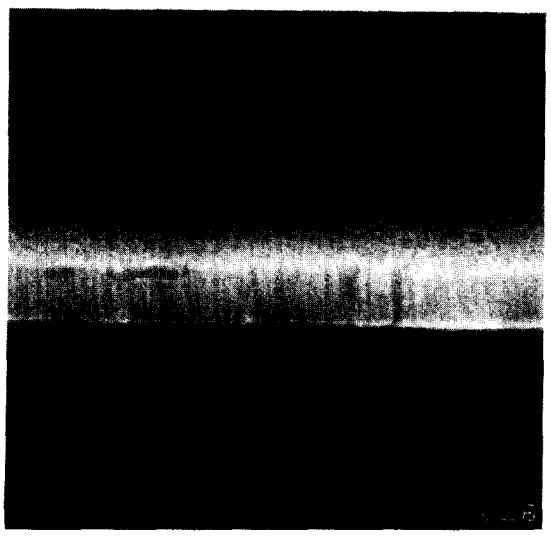

Fig. 3. SEM micrograph of a ZnO/GaAs fractured film showing a dense columnar structure.

The rocking curves measured for the (002) lines are shown in Fig. 2. Again the ZnO/SiO_2/GaAs sample exhibits the best result, with a FWHM of only 2.18°, a value which compares very favourably with the values of 2.5–3.0° usually reported for other techniques [19, 20] and is similar to those of our ZnO films grown by PLD on Si or glass substrates [13, 14]. The standard deviation of the rocking curve is $\sigma = 0.93°$, a value usually measured only for ZnO films deposited on sapphire substrates [19].

SEM investigations showed relatively smooth surfaces for the deposited films with very few droplets being present. The fractured ZnO film surfaces reveal a dense, columnar structure as shown in Fig. 3, similar to that seen for ZnO films grown on Si by either PLD or r.f. sputtering.

3. Conclusions

In conclusion, some of the best quality ZnO films in terms of crystalline structure have been grown on GaAs substrates at substrate temperatures around 350 °C employing the PLD method. The SiO_2 buffer layer improves significantly the crystallinity of the ZnO film and reduces the built-in strain. Under optimised conditions, highly c-axis oriented ZnO films having a FWHM value of the (002) XRD reflection line of less than 0.13° and a FWHM value of the rocking curve of the (002) line of only 2.18° can be grown. The surface morphology has been found to be smooth while the fractured surface exhibited a dense, columnar structure.

Acknowledgements

This work was funded by SERC contract No. GRH67133. We wish to thank Dr. P. Bergonzo for help with the SiO_2 buffer layer photodeposition and Prof. M. Green for the use of the ellipsometer. Additional

funding was also provided by the scientific collaboration between the Netherlands Organization for Scientific Research (NWO) and the British Council – Prog. No. JRP006.

References

[1] O. Madelung (ed.), *Landolt-Bornstein New Series*, Vol. 17b, Springer–Verlag, Berlin, 1982.
[2] E. Kaldis (ed.), *Current Topics in Materials Science*, Vol. 7, North-Holland, Amsterdam, 1981.
[3] T. Minami, H. Sonohara, S. Takata and H. Sato, *Jpn. J. Appl. Phys., 33* (1994) L743.
[4] D. H. Zhang and D. E. Brodie, *Thin Solid Films, 238* (1994) 95.
[5] S. Kohiki, M. Nishitani, T. Wada and T. Hirao, *Appl. Phys. Lett., 64* (1994) 2876.
[6] H. Sato, T. Minami, Y. Tamura, S. Takata, T. Mouri and N. Ogawa, *Thin Solid Films, 246* (1994) 86.
[7] P. F. Carcia, Z. G. Li and H. W. van Kesteren, *Thin Solid Films, 246* (1994) 126.
[8] T. Itoh and T. Suga, *Appl. Phys. Lett., 64* (1994) 37.
[9] H. K. Kim and M. Mathur, *Appl. Phys. Lett., 61* (1992) 2524.
[10] W.-C. Shih and M.-S. Wu, *J. Cryst. Growth, 137* (1994) 319.
[11] Y. Kim, W. D. Hunt, F. S. Hickernell and R. J. Higgins, *J. Appl. Phys., 75* (1994) 7299.
[12] H. K. Kim, W. Kleemeier, Y. Li, D. W. Langer, D. T. Cassidy and D. M. Bruce, *J. Vac. Sci. Technol., B12* (1994) 1328.
[13] S. Amirhaghi, V. Craciun, F. Beech, M. Vickers, S. Tarling, P. Barnes and I. W. Boyd, *Mater. Res. Soc. Proc., 285* (1993) 489–494.
[14] V. Craciun, D. Craciun, J. Elders, J. G. E. Gardeniers and I. W. Boyd, presented at E-MRS'94 Spring Meeting, to be published in *Appl. Surf. Sci.*
[15] P. Bergonzo, P. Patel, I. W. Boyd and U. Kogelschatz, *Appl. Surf. Sci., 54* (1992) 424.
[16] P. Bergonzo, I. W. Boyd and U. Kogelschatz, *Appl. Surf. Sci., 69* (1993) 393.
[17] *Powder Diffraction File no. 36–1451*, Joint Committee on Powder Diffraction Standards, 1986.
[18] B. D. Cullity, *Elements of X-ray Diffraction*, Addison-Wesley, Reading, MA, 1978.
[19] Y. Igasaki and H. Saito, *J. Appl. Phys., 70* (1991) 3613.
[20] F. C. M. van de Pol, F. R. Blom, and Th. J. A. Popma, *Thin Solid Films, 204* (1991) 349.