Growth Kinetics Study of Pulsed Laser Deposited ZnO Thin Films on Si (100) Substrate

Deepak N. Bankar[1], Suhas M. Jejurikar[2], K. P. Adhi[1], A. V. Limaye[1] and A. G. Banpukar[1]

[1]*Center for Advanced Studies in Materials Science and Condensed Matter Physics, Department of Physics, University of Pune, Pune - 411 007, India*
[2]*Department of Electrical Engineering, Indian Institute of Technology, Bombay, Powai, Mumbai - 400 076, India*

Abstract. Surface morphology and kinetic roughness of ZnO thin films grown on Si(100) substrate for different time durations by pulsed laser deposition technique were analyzed using Atomic Force Microscopy images. Dynamic scaling approach was used for quantitatively analyzing the surface topology in terms of height difference correlation function $G(r, t)$ and interface width $w(t)$. Dynamic scaling approach is used to find roughness exponent α and growth exponent β, which yields the values 0.67 and 0.49 respectively. This α value is in good agreement with the value predicted by surface diffusion driven model. However, value of growth exponent β is higher than the model value. The deviation suggests anisotropy in film growth. X-ray diffraction data shows the preferential *c*-axis growth supporting deviation from the diffusion driven model.

Keywords: Dynamic Scaling, Pulsed Laser Deposition, AFM, ZnO thin films.
PACS: 68.35.Ct; 81.15.Fg; 68.37.Ps; 68.55.j

INTRODUCTION

In recent years, several theoretical and experimental studies [1,2] on the kinetic surface roughening in growth of solid film have revealed the simple scaling relation in term of the height-difference correlation function $G(r,t)$ defined as:

$$G(r,t) = \left\langle \left(h(x,y) - h(x',y') \right)^2 \right\rangle$$

where, $h(x, y)$ is the height of the surface at the in-plane coordinates (x, y) and

$$r = ((x-x')^2 + (y-y'))^{1/2}$$

The $G(r, t)$ of random self-affine fractal surface contains at least three important parameters: the vertical correlation length or interface width $w(t)$, the lateral correlation length $\xi(t)$ and the roughness exponent α, which are defined as below:

$$G(r,t) \propto \begin{cases} Cr^{2\alpha}, & r << \xi(t) \\ 2w^2(t), & r >> \xi(t) \end{cases}$$

Here, $\xi(t)$ provides a length scale, which distinguishes the short-range and long-range behaviors of the rough surface. It is the lateral correlation length that gives an average measure of the lateral coarsening size at the growth time (or sample thickness) t, and it is the distance within which the surface variations are correlated. The interface width $w(t)$ describes the surface roughness along the vertical direction during the growth process and it is defined as the root mean square (rms) surface height fluctuations, which will evolve with time in the form of power laws $w(t) \propto t^{\beta}$, where β is the growth exponent. Both $w(t)$ and $\xi(t)$ are can be used for statistical description for the global surface morphology. The roughness exponent α is an important parameter to describe self-affine fractal surface and it describes the local surface roughness. A small value of α (< 0.5) corresponds to a short-range surface, while large value of α (>0.5) corresponds to more jagged local surface morphology [3,4].

In this paper, using atomic force microscopy (AFM) images, we have investigated in detail, the surface roughening in the pulsed laser deposited (PLD) ZnO thin films grown on Si (100) substrate.

EXPERIMENTAL

A ZnO films were deposited on Si(100) substrate by pulsed laser ablation of ZnO target for different time duration (3, 5, 10,15 25 min) in an oxygen

Optics: Phenomena, Materials, Devices, and Characterization
AIP Conf. Proc. 1391, 101-103 (2011); doi: 10.1063/1.3646792

ambient. The depositions were carried out at optimized O_2 pressure and laser energy density which is discussed elsewhere [5]. Profilometer is used to estimate the film thickness, which is found to be 100, 180, 350, 550 and 900 Å respectively. The films surface topography was then studied using AFM, in contact mode. The films were scanned over the area of 10×10 to 0.1×0.1 μm^2. For the present study, images of 1×1 μm^2 were used for analysis. Each image employs a pixel size of 512×512 and height of each pixel is represented in 256 grey levels. For a substantial statistical averaging, seven images of each sample from different areas were randomly selected. A standard procedure has been adopted to analyze the AFM images for scaling [1,6].

RESULTS AND DISCUSSION

AFM images (Fig. 1(a-d)) show the surface morphology of ZnO thin films with increase in film thickness. Interestingly, it is observed that in the initial stages average mound size increases. However, in case of film thickness 350 Å, average mound size is observed to be minimum.

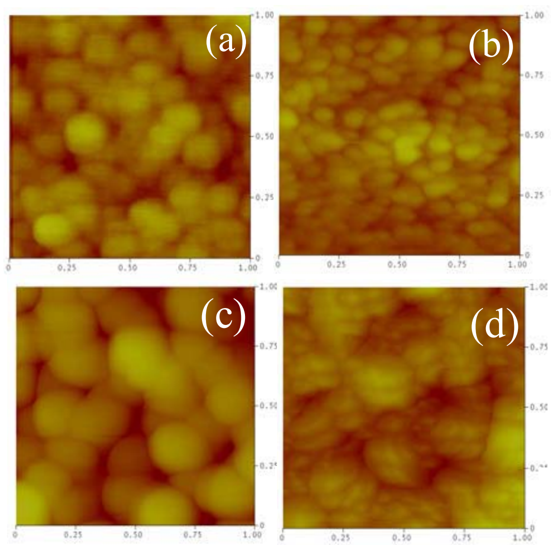

FIGURE 1. AFM images of ZnO films deposited for thickness (a) 100, (b) 350, (c) 550 and (d) 900 Å

The data of the height-difference correlation function $G(r, t)$ is obtained by processing AFM images for different thickness t. Figure 2 shows the variation in $G(r, t)$ plotted against r on log-log scale. For the short range ($r << \xi(t)$), linear relationship between $G(r, t)$ and r is observed, corresponding to the proportionality of $G(r, t) \propto r^{2\alpha}$, the slope of the linear part in the curve is 2α, and it can be obtained by the linear fit to the data. At sufficiently large r ($r >> \xi(t)$), $G(r, t)$ tends to be a constant value of $2w(t)^2$.

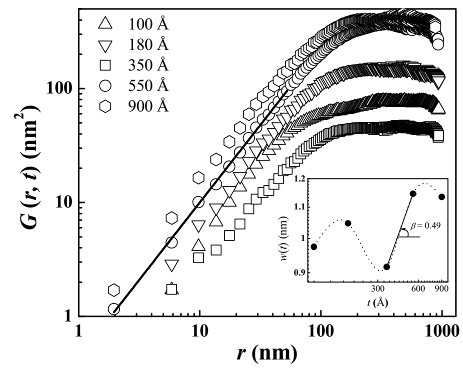

FIGURE 2. Height-difference correlation function $G(r, t)$ as a function of r plotted on log-log scale. Inset shows plot of interface width $w(t)$ against film thickness t on log-log scale.

The values of scaling exponents such as, α, $w(t)$ and $\xi(t)$ for the ZnO films are listed in Table 1. The roughness exponent α is found to be 0.67±0.06. The turning point in the curve determines the lateral correlation length $\xi(t)$. In order to obtain the values of $\xi(t)$, we fit the curves of $G(r, t)$ at a specific thickness t by the phenomenological function proposed by Sinha et al. [7]:

$$G(r,t) = 2w^2 \left\{1 - \exp\left(-(r/\xi)^{2\alpha}\right)\right\}$$

TABLE 1. Surface parameters α, w(t) and ξ(t)

Thickness *t* Å	α	*w*(t) nm	ξ(t) nm
100	0.78	0.97	94.45
180	0.65	1.05	123.05
350	0.60	0.92	107.42
550	0.67	1.15	154.30
900	0.67	1.14	138.67

The plot of roughness w(t) versus thickness t is shown in Fig. 2 (inset). It can be seen from the figure that relation of w(t) against t can be divided into two stages. When the film thickness is shorter than 350 Å, the value of w might be related to the overall influence by the factors of random fluctuations and the diffusion process of the deposition particles, the lateral strain, and the effect due to substrate-film lattice mismatch might be responsible for the initial roughening. The roughness w decreases with thickness (t < 350 Å) and the surface fluctuations became small, which indicates that, the presence of surface diffusion smoothes the surface roughness in the early stages of film growth. For the thickness $t \geq 350$ Å the dynamics of the surface morphology is different and the process of the formation of mounds starts after this thickness therefore the roughness w increases with further growth, and it is proportional to t^{β}. The data of roughness w versus thickness t for $t \geq 350$ Å to the relation of $w(t) \propto t^{\beta}$ and the fit result gives $\beta = 0.49$.

Fractal dimension $d_F = 3-\alpha$ [8] of the ZnO thin surface is calculated from the roughness exponents found to be 2.33 ± 0.06, which is higher than d_F for plane surface leading to the conclusion that the growth is dominated in vertical direction. The value obtained for dynamic scaling exponent z using relation $\xi \sim t^{1/z}$ and $z = \alpha/\beta$ ($z = 1.37$) do not match [9]. This clearly indicates anomalous scaling holds in case of ZnO.

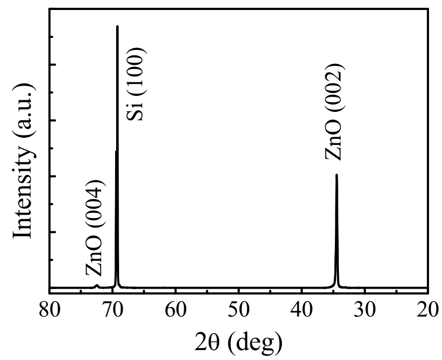

FIGURE 3. XRD patterns for ZnO film for film thickness t =550 Å

The value of α is greater than 0.5 as predicted by KPZ [1]. But, this value is in good agreement with that predicted by surface diffusion-driven growth model [10]. The α value indicates that diffusion of atoms on the substrate. The growth exponent β value is higher than that predicted by surface diffusion driven model value 0.333 [1], which can be explained on the basis of existence of edge barrier such as Schwoebel effect [11-14], where the diffusing atoms has tendency to ascend the step on the diffused atom rather than diffusing on the substrate. This introduces asymmetry in the diffusion process leading to the growth along preferred directions [12, 15]. To understand the origin of high value of β we characterized the films structurally using X-ray diffraction technique (Fig.3). Highly oriented growth of ZnO along c-axis is confirmed due to existence of (002) and (004) peaks. Thus further confirms that ZnO grows via asymmetrical surface diffusion of atomic species.

CONCLUSION

In summary, the scaling exponents for the Pulsed Laser deposited ZnO thin films are investigated from the AFM images. The roughness exponent α agrees with the value supported by the surface diffusion driven growth model while the model does not support the values of β. From fractal dimension value, shows vertical (columnar) growth on Si substrate, leading to conclusion that the growth kinetics is completely different.

ACKNOWLEDGMENTS

The authors would like to thank Dr. V. Ganesan, UGC-DAE Consortium for Scientific Research for providing AFM facility and Mr. Mohan Gangrade and Ms. Deepti Jain for their assistance in measurements.

REFERENCES

1. A. L. Barabasi and H. E. Stanley, "Fractal Concepts in Surface Growth", Cambridge University press, Cambridge, England, 1995.
2. "Dynamics of Fractal Surfaces", edited by F. Family and T. Vicsek. World Scientific, Singapore, 1991; C. V. Dharmadhikari, A. O. Ali, N. Suresh, D. M. Phase, S. M. Chaudhari, V. Ganesan, A. Gupta and B. A. Dasannacharya, *Solid State Commu*. **114**, 377-381 (2000).
3. Yun-Yan Liu, Chuan-Fu Cheng, Shan-Ying Yang, Hong-Sheng Song, Gong-Xiang Wei, Cheng-Shan Xue, Jun-Cheng Liu, *Thin Solid films*, doi: 10.1016/j.tsf.2011.02.056 (2011).
4. H. N. Yang, Y.P. Zhao, G. C. Wang, T. M. Lu, *Phys. Rev. Lett.* **76**, 3774-3777 (1996).
5. Suhas M. Jejurikar et.al, *J. Appl. Phy.,* **99**, 014907-014914 (2006).
6. S. M. Jordon, R. Schad, J. F. Lawler, D. J. L. Herrmann and H. van Kempen, *J. Phys: Cond. Matt.* **10**, L335-L358 (1998).
7. S. K. Sinha, E. B. Sirota, S. Garoff, *Phys. Rev. B.* **38**, 2297-2311 (1998).
8. B. Mandelbrot, "The Fractal Geometry of Nature" Freeman, New York, 1982.
9. M. Saitou, K. Hamaguchi and W. Oshikawa, *J. Electrochemical Society*, **150**, C99-C103 (2003).
10. Z. W. Lai and S. D. Sarma, *Phys. Rev. Lett.* **66**, 2348-2351 (1991).
11. R. L. Schwoebel and E. J. Shipsey, *J. Appl. Phys.*, **37**, 3682-3686 (1966).
12. H. J. Ernst, F. Fabre, R. Folkerts and J. Lapujoulade, *Phys. Rev. Lett.* **72**, 112-115 (1994).
13. L. Vazques, J. M. Albella, R. C. Salvarezza, A. J. Arvia, R. A. Levy and D. Perese, *Appl. Phys. Lett.*, **68**, 1285-1287 (1996).
14. M. Saitou, W. Oshikawa, M. Mori and A. Makabe, *J. Electrochemical Society*, **148**, C780-C783 (2001).
15. H. Park, A. Provata and S. Render, *J. Phys. A.*, **24**, L1391-L1397 (1991).