In situ x-ray diffraction study of PLD growth of ZnO-Al_2O_3

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Introduction

Pulsed-laser deposition (PLD) provides epitaxial film and heterostructure growth conditions that result in materials properties not available by near-equilibrium growth methods. ZnO is a technologically important material of strong interest both as a wide band-gap semiconductor and as a buffer layer for lattice-matched growth of GaN films. We have constructed a PLD film growth chamber for *in situ* time-resolved x-ray diffraction investigations of the deposition and crystallization processes during growth of thin films using the UNI-CAT undulator beamline at the Advanced Photon Source. In this report, we discuss initial film growth measurements investigating heteroepitaxial PLD growth of ZnO on Al_2O_3.

Methods and Materials

The laser-ablation film-growth chamber is mounted on a so-called 2 + 2 x-ray diffractometer providing for surface-scattering measurements in specular and off-specular directions. A rotatable mirror is used to reflect ~ 0.5–1.0 J, 25 ns excimer laser pulses onto the ablation target as the chamber is rotated for surface diffraction measurements. The sapphire substrates were <0001> oriented wafers that had been oxygen annealed at high temperature. Specular and off-specular truncation-rod intensity measurements were performed. Static *in situ* measurements were performed after varying depositions to monitor the ZnO in-plane lattice parameter relative to the ~17% smaller lattice parameter of the Al_2O_3 substrate.

Results

Specular CTR anti-Bragg measurements at the sapphire (0 0 5/2) position showed only one well-defined growth oscillation, indicating three-dimensional growth rather than layer-by-layer growth. Off-specular CTR measurements along the (H H 0.3) direction showed thermally activated relaxation of the 17% lattice mismatch between ZnO and Al_2O_3 along with a 30º in-plane rotation around the c-axis. As shown in Fig. 1, a broad, nearly relaxed ZnO in-plane diffraction peak appears after the deposition of three monolayers at 400º C (~25 pulses/monolayer), while a sharper and more fully relaxed ZnO peak appears after only two monolayers at 585º C. Subsequent measurements (not plotted here) show that incommensuration occurs within the first layer of deposition but with substantially compressed in-plane ZnO lattice parameters resulting from the underlying sapphire substrate. We note from Fig.1 that full relaxation occurs only after several layers are deposited and that relaxation occurs more quickly at the higher temperature.

The in-plane diffraction widths in Fig. 1 correspond to in-plane coherence sizes for the ZnO films of ~75–100 Å, compared to lateral sizes of ~150–200 Å we have observed by atomic force microscopy measurements made on these samples. This result indicates the presence of structural defects within individual three-dimensional clusters formed in the first few layers.

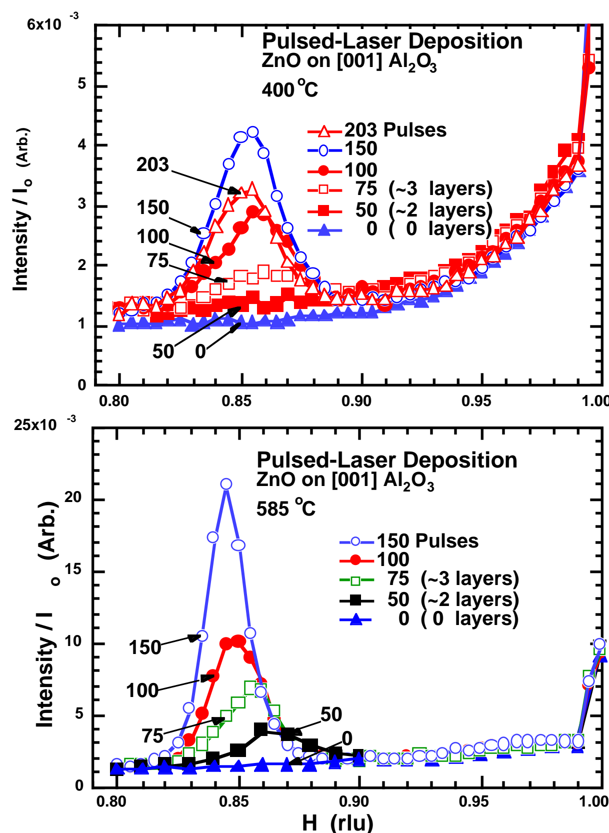

Figure 1: CTR x-ray diffraction measurements along the (H H 0.3) direction (sapphire units) indicating temperature dependent relaxation. H = 0.83 corresponds to full relaxation.

Discussion

The observation of full lattice relaxation of ZnO after a few layers is consistent with Transmission Electron Microscopy observations of high densities of dislocations at the ZnO-Al_2O_3 boundary [1] that decrease rapidly with distance from the interface. More detailed measurements at additional temperatures and varying deposition conditions are in progress to investigate ZnO film growth on ZnO as well as sapphire substrates.

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Reference

[1] J. Narayan, K. Dovidenko, A.K. Sharma, and S. Oktyabrsky, *J. Appl. Phys*. **84**, 2597 (1998).