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Lateral epitaxial overgrowth of ZnO layers on hexagonally patterned buffer layers in low-temperature aqueous solution

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ABSTRACT

In conjunction with lift-off patterning and thermal annealing, we have fabricated hexagonal arrays of epitaxial $ZnAl_2O_4$ buffer layers on sapphire substrates. Taking advantage of the small lattice mismatch between ZnO (0001) and $ZnAl_2O_4$ (111), site-controlled epitaxial growth of ZnO prisms occurred on the pre-patterned, lattice-matched buffer layers. Subsequently, using a continuous flow reactor, long-duration maskless lateral epitaxial overgrowth (LEO) of ZnO prisms led to coalescence into continuous, thick layers within aqueous solutions at low temperature. X-ray diffraction (XRD) confirmed that LEO on the hexagonally patterned buffer layers facilitated the fabrication of strain-relieved, wing-tilt-free, thick ZnO layers. The dislocation density at the coalesced LEO-grown ZnO layer was approximately 10^8 cm^{-2}. The spatial variation in the micro-photoluminescence characteristics of the LEO-grown layer suggested that the improvement in crystalline quality occurred in the overgrown wings, originating from the decrease in dislocation in the ZnO layer during the LEO process.

1. Introduction

Zinc oxide (ZnO) is a promising material for use in optoelectronic devices because of its attractive optoelectronic properties, including a direct band gap of 3.3 eV at 300 K, a high exciton binding energy (61 meV), and high visible transparency [1]. In addition, the room temperature ferromagnetic behavior was discovered in pure nano-grained ZnO films [2]. The growth of high-quality ZnO epitaxial layers is necessary for the many applications of ZnO-based optoelectronic devices. Several techniques have been developed for the growth of epitaxial ZnO layers, including metal–organic chemical vapor deposition (MOCVD) [3], pulsed laser deposition (PLD) [4], molecular beam epitaxy (MEB) [5], sputtering [6], atomic layer deposition [7], and hydrothermal growth [8]. Among them, hydrothermal growth is particularly attractive for the preparation of ZnO epitaxial layers and single-crystalline nanostructures because of the low growth temperature (<90 °C) and the use of simple facilities [8,9]. However, the growth of ZnO epitaxial layers having low defect densities remains a considerable challenge because of large lattice mismatch.

Lateral epitaxial overgrowth (LEO) is the most promising method for decreasing defects as demanded for device applications. Andeed et al. were the first to demonstrate the LEO of ZnO layers of low dislocation density (2×10^8 cm^{-2}), from aqueous solutions at 90 °C and using a patterned photoresist as a mask layer for LEO processing [10]. Since then, various mask-based and maskless LEO approaches have been investigated for the preparation of ZnO epilayers with low dislocation density [11–15]. In a previous study, we developed a facile approach, using a stripe-patterned buffer layer, for the maskless LEO of ZnO layers having low dislocation. However, a slight wing tilt, at an angle of 0.1°, remained in the LEO ZnO layer [13]. Nevertheless, when using a conventional autoclave vessel to implement the long-duration hydrothermal growth, the concentration of soluble zinc species in the growth solution was insufficient to achieve full coalescence of the LEO ZnO layer. To facilitate coalescence of the ZnO layer, the LEO process had to be interrupted to refresh the growth solution, increasing the risk of contamination. In this present study, we employed a continuous flow reactor that maintained the concentration of zinc species in solution during crystal growth by continuously delivering the precursor solution, thereby facilitating long-duration LEO. Comparing with the previous work (stripe pattern), we exploited the hexagonal array of the $ZnAl_2O_4$ buffer layer to perform maskless LEO of the ZnO layer. The influence of hexagonally patterned architecture on the reduction in defects and wing tilts in LEO-grown ZnO layer was investigated in this work.

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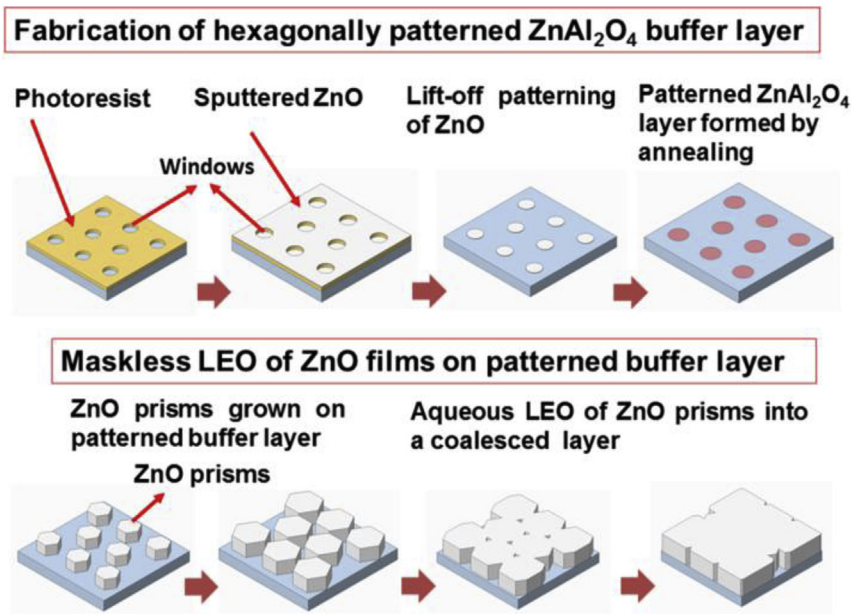

Fig. 1. Schematic representation of the process flow for the fabrication of a patterned buffer layer and for the LEO of a ZnO film on the patterned buffer layer.

2. Experimental procedure

The process flow for fabrication of the hexagonally patterned buffer layer on a sapphire substrate and the LEO of the ZnO layer is illustrated in Fig. 1. The patterned photoresist having a hexagonal array of holes was generated on $(11\bar{2}0)$ plane sapphire substrates using conventional photolithography. The exposed regions (holes) are called "windows"; the regions covered by the photoresist are called "wings." Following deposition, through sputtering, of a thin ZnO coating over the whole area of the patterned photoresist sample, the patterned epitaxial $ZnAl_2O_4$ buffer layer was formed on the sapphire substrate by combining lift-off and annealing processes. $ZnAl_2O_4$ layers in epitaxial registry with underlying sapphire substrates $((111)_{ZnAl2O4}||(11\bar{2}0)_{Al2O3}$ and $[1\bar{1}2]_{ZnAl2O4}||[0001]_{Al2O3})$ are readily formed after annealing of ZnO-coated sapphire samples [8,16]. The detailed fabrication procedure of epitaxial patterned $ZnAl_2O_4$ buffer layer on sapphire substrates by combining lift-off and thermal annealing processes can be referred previous works [13,16]. Prior to site-controlled epitaxial growth of ZnO, the annealed sample was soaked in dilute HCl to remove any residual ZnO grains on the sapphire substrate. Site-controlled epitaxial growth of ZnO prisms on the patterned buffer layer was performed by immersing it in a 200 mL aqueous solution of 0.1 M zinc nitrate hexahydrate [$Zn(NO_3)_2{\cdot}6H_2O$, 99%, Showa],

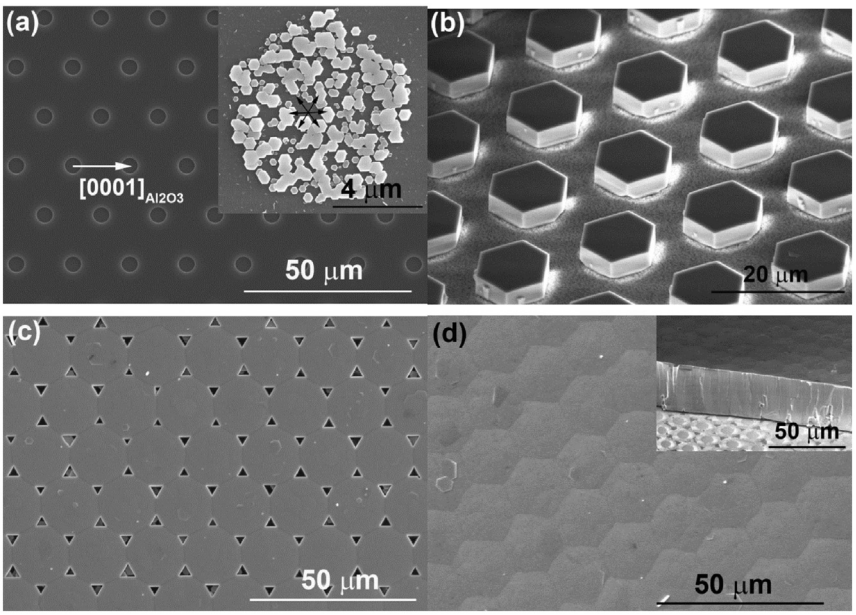

Fig. 2. (a) SEM image of a photoresist layer having a hexagonal array of circular holes; inset: site-controlled epitaxial growth of tiny ZnO crystals on the window regions during the early stages of growth. (b) Site-controlled epitaxial growth of ZnO prisms on the patterned buffer layer. (c, d) Long-duration aqueous LEO of ZnO for (c) 60 and (d) 84 h; inset to (d): tilted-view SEM image of a cleaved cross-section of the fully coalesced LEO-grown ZnO layer (84 h).

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