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Surface chemistry and growth mechanisms studies of homo epitaxial (1 0 0) GaAs by laser molecular beam epitaxy

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ABSTRACT

In this paper, GaAs thin film has been deposited on thermally desorbed (1 0 0) GaAs substrate using laser molecular beam epitaxy. Scanning electron microscopy, *in situ* reflection high energy electron diffraction and *in situ* X-ray photoelectron spectroscopy are applied for evaluation of the surface morphology and chemistry during growth process. The results show that a high density of pits is formed on the surface of GaAs substrate after thermal treatment and the epitaxial thin film heals itself by a step flow growth, resulting in a smoother surface morphology. Moreover, it is found that the incorporation of As species into GaAs epilayer is more efficient in laser molecular beam epitaxy than conventional molecular beam epitaxy. We suggest the growth process is impacted by surface chemistry and morphology of GaAs substrate after thermal treatment and the growth mechanisms are discussed in details.

1. Introduction

Preparation of high quality epitaxial semiconductor layers is very important for device technology [1]. III–V compound semiconductors are widely used in high-speed, low-noise solid state devices and optoelectronics [2]. Especially, GaAs has attracted great attention for fabrication of high electron mobility transistors [3]. GaAs-based compound materials, such as AlGaAs, and its different alloy compositions play an important role for terahertz quantum cascade laser [4] and light emitting diode [5]. In order to fabricate high performance devices, the fundamental understanding of material growth processes is critical, and is the aim of many researches.

There are many reports on the growth mechanisms of GaAs epilayer by metal organic chemical vapor deposition (MOCVD) and molecular beam epitaxy (MBE) [6–9]. Laser molecular beam epitaxy (LMBE) is a pulsed laser thin film deposition technique, which is a relatively new growth technique that has been applied to a wide range of materials [10]. There are several advantages of LMBE for depositing high-quality thin films and make it worthy of study as a good method of growing III–V compound semiconductors. For LMBE, the congruent ablation achieved with short laser pluses allows deposition of a multicomponent material by employing a single target. The growth rate can be varied via adjusting the repetition rate of the laser, which is useful for both atomic level investigations and thick layer growth. Moreover, the peak mass arrival rate of LMBE is significant higher than the steady state MBE flux required for the same growth rate. The adsorbed species may also have a significant higher kinetic energy in LMBE, creating the potential for smoother film morphology at low substrate temperatures [11]. But up to now, only a few studies have been focused on the growth mechanisms of homo epitaxial (1 0 0) GaAs by using pulsed laser deposition technique [12]. Using atomic force microscopy (AFM) and reflection high energy electron diffraction (RHEED), Pun et al. [12] studied the surface morphology of homo epitaxial GaAs by pulsed laser deposition technique. However, the changing of surface chemistry during growth process is still not clear. In this study, we present a detailed investigation on the surface chemistry during growth process using *in situ* X-ray photoelectron spectroscopy (XPS). The growth mechanisms of GaAs epilayer are discussed based on the surface chemistry and morphology evolution.

2. Experimental details

The deposition of GaAs thin film was achieved using an ultra-high vacuum LMBE system, in which a KrF laser ($\lambda = 248$ nm) pulsed at 1 Hz was incident on a 99.999% GaAs target with a fluence of 2 J/cm^2, resulting in a highly energetic plume that subsequently deposited on a (1 0 0) GaAs substrate located ~6 cm from the target.

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Before deposition, GaAs substrate had been treated at 600°C for ~30 min to desorb the oxides from the substrate. The epitaxial GaAs layer was grown at the same temperature with a base pressure better than 5×10^{-7} Pa.

The surface morphologies of GaAs substrate and epilayer were characterized by a FEI model Nova 600i scanning electron microscopy (SEM). *In situ* XPS investigation was carried out in the analysis chamber with a base pressure better than 2×10^{-7} Pa. The Al Kα (1486.6 eV) monochromatized radiation was used in this study. After growth, transmission electron microscopy (TEM) investigation was carried out using a FEI Tecnai 20ST and the cross-section specimen was prepared by focused ion beam (FIB). High resolution grazing incident X-ray diffraction (GIXRD) measurements were performed at the U7B beamline of National Synchrotron Radiation Laboratory of China (NSRL) using a wavelength of 0.1547 nm at the 0.2° angle of incidence to the specimen surface.

3. Results

Fig. 1(a) shows the surface morphology of GaAs substrate before thermal treatment. It can be seen from the image that the surface of GaAs substrate is very smooth. But a high density of pits on the surface is observed after thermal treatment as shown in Fig. 1(b). Eventually, the pits diminish dramatically to form a continuous film as shown in Fig. 1(c) after 60 min of growth.

The RHEED images, with typical patterns of the pregrowth substrate surface and film growth for 10 and 25 min are shown in Fig. 2. The pattern of GaAs substrate before deposition as shown in Fig. 2(a), compared to other patterns, clearly shows larger broad diffraction spots. It is believed that the multiple diffraction spots indicate partial transmission diffraction occurring through the protuberances with the presence of small vicinal disordered domain on GaAs substrate surface [13], which also can be confirmed by the SEM image as shown in Fig. 1(b). After 10 min of growth, the diffraction pattern as shown in Fig. 2(b) shows an elongated and narrowed diffraction spots and this evolution in RHEED pattern indicates a surface morphology transition from an extremely rough surface to a smoother surface. Fig. 2(c) shows the RHEED pattern after 25 min of growth, and the diffraction pattern does not change significantly till the end of growth, indicating the surface morphology of sample is stable after 25 min of growth. This result is different from Pun et al. [12] who observed a strong Ga-rich (4 × 6) surface symmetry during the growth process, the difference is ascribed to the different experimental conditions.

In situ XPS spectra were taken to gain insight of the surface chemical composition change during film growth. Photoelectron peaks were recorded and resolved by spectrum synthesis in which the spectral line shapes were simulated by curve fitting with a product of Gaussian and Lorentzian lines. Fig. 3 shows the Ga 3d core level spectra at different time of the deposition process. These spectra can be decomposed into two components which corresponding to Ga(–As) bond at the binding energy of ~19.3 eV and Ga–Ga bond at ~18.7 eV [14], besides we cannot observe any other bonding states in Ga 3d core level. Fig. 3(a) shows Ga 3d core level spectra after thermal treatment, and the peak area ratio value of Ga(–As) bond and Ga–Ga bond is ~0.66. Whereas the peak area ratio goes up to ~1.08 after 10 min of growth as shown in Fig. 3(b). Decomposition of the core level spectra indicates more Ga–Ga bonds have converted to Ga(–As) bond after 20 min of growth as shown in Fig. 3(c), and the peak area ratio increases to ~1.17. Fig. 3(d) shows Ga 3d core level spectra after 30 min of growth, the value of peak ratio is ~1.24 which is a little larger than the value for 20 min of growth. A surprising change has been observed in Fig. 3(e), showing Ga 3d core level spectra after 60 min of growth. The peak area

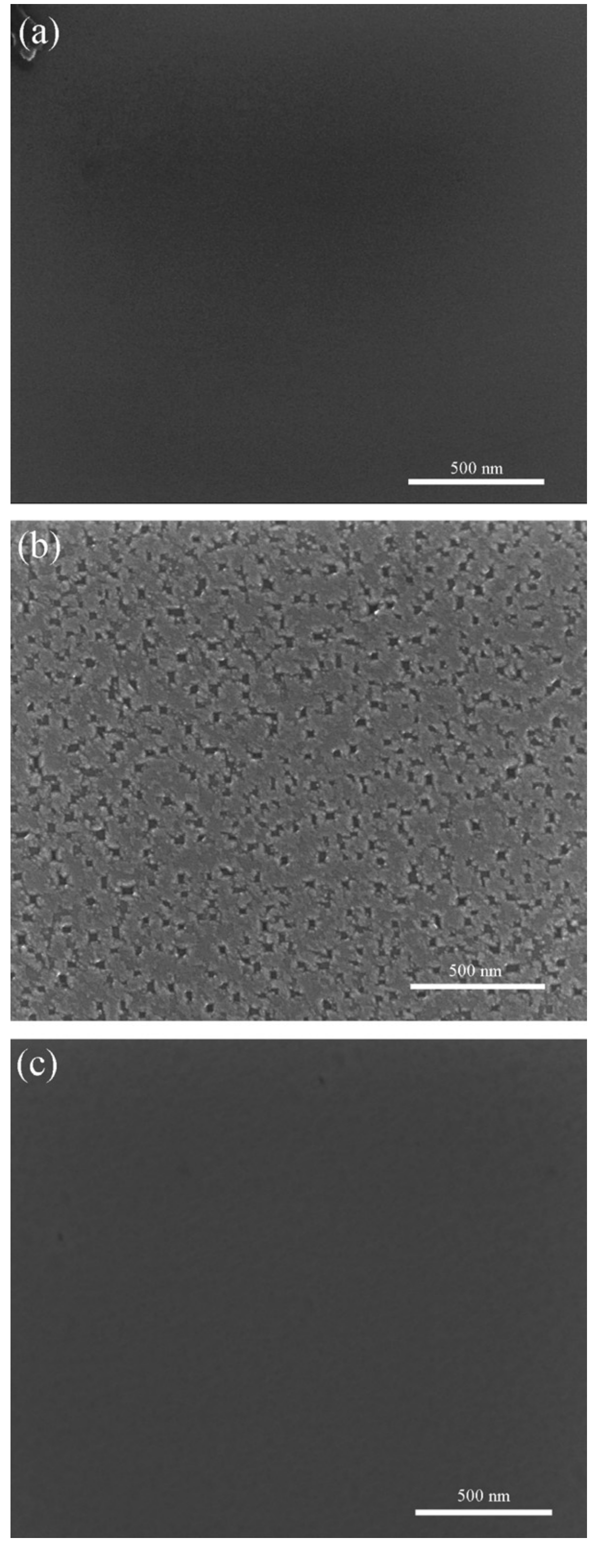

Fig. 1. SEM images of GaAs substrate and epilayer surface: (a) a bare (1 0 0) GaAs substrate, while (b) was GaAs substrate taken after thermal treatment and (c) GaAs epilayer taken after 60 min of growth.

of Ga(–As) bond is much larger than Ga–Ga bond, which almost makes the whole contribution to Ga 3d core level spectra, and the peak area ratio value sharply increases to ~8.21.

The Ga:As atomic concentration ratios, which are calculated by measuring the area under Ga 3d and As 3d photoelectron spectral lines after subtracting a Shirly background, reveal the contribution