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ScienceDirect
Journal of Crystal Growth 307 (2007) 19–25
JOURNAL OF CRYSTAL GROWTH
www.elsevier.com/locate/jcrysgro

Influence of In on the surface morphology of HVPE grown GaN

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Received 23 August 2006; received in revised form 4 June 2007; accepted 4 June 2007
Communicated by D.W. Shaw
Available online 9 June 2007

Abstract

In this paper, a study is presented on the effect of In on the surface morphology of GaN grown by HVPE. Experiments are performed with N_2 and H_2 as the carrier gasses, both with and without In present in the reactor. The adding of In increases the morphological quality of the grown layers; this effect is most strongly observed for N_2 as the carrier gas. It is found that adding In reduces the growth rate and also increases the steepness of the growth hillocks on the surface. The step velocity, which is calculated from hillock slopes and the growth rate, decreases upon adding In. Without In, trails are visible across the surface where steps are distorted by passing a dislocation outcrop. With In present this pinning is still present, however, the trails do not form. Two possible explanations for this phenomenon are an increased surface diffusion due to a mono- or bi-layer of In on the surface and the slower step motion when In is present.

PACS: 68.55.Jk

Keywords: A3. Hydride vapour phase epitaxy; B1. Nitrides

1. Introduction

In many systems of semiconductor growth the use of isoelectronic doping is shown to improve the quality of the layers. Especially the use of In and InAs for GaAs and InP is well known for improving the crystalline quality of the layer [1]. Ideally, the isoelectronic doping improves the morphology and does not get incorporated into the crystal lattice, i.e. it behaves as an ideal surfactant. For GaN, such an effect of improving quality has been shown for the addition of In during the growth by both metal-organic chemical vapour deposition (MOCVD) and molecular beam epitaxy (MBE) [2–6].

In MBE the addition of In to the system results in more uniform growth [2], and in the improvement of the morphological and optical properties of the grown GaN layer. It is stated that In acts as a surfactant, modifying the surface kinetics and increasing the Ga mean free path at the surface, allowing 2D growth. Indium is an interesting material to be used as a surfactant. The weaker In–N bond as compared with the Ga–N bond is the main driving force for surface segregation of In and little In will be incorporated: no alloying will take place. The same effects have been observed in MOCVD growth. Experiments with In as surfactant for MOCVD growth of GaN at 950 °C showed that In was incorporated at a level less than 0.001% [7]. Further, the surface morphology as well as the optical properties improve [3–5]. Like in MBE, this is attributed to In modifying the growth kinetics [4,6]. In addition, the dislocation density reduces and the formation of deep energy levels is suppressed [8,9]. Both in MBE and MOCVD the addition of In to the growth system leads to better quality layers. In a theoretical paper [6], this is explained by the formation of an In mono- or bi-layer at the surface. This layer facilitates the diffusion of N over the surface and leads to better quality material. These calculations were performed for a substrate temperature of 827 °C, which is a lower temperature than used for MOCVD growth. However, in Ref. [3] experimental

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doi:10.1016/j.jcrysgro.2007.06.005

evidence of In acting as a surfactant is shown for the higher temperature of 1100 °C.

On the effect of isoelectronic doping of GaN with In in the HVPE growth of GaN, to our best knowledge no publications are available. The growth of pure InN by HVPE has been tried. Because of the high dissociation pressure of InN only growth at low temperatures is possible. Depending on the choice of precursors (InCl or $InCl_3$), the growth temperature for pure InN varies from 530 to 750 °C [10–13]. In order to grow alloys of InGaN, the growth temperature also needs to be low. This does not promote obtaining good quality layers [14], moreover, still only very little In can be incorporated. Besides the temperature also the carrier gas is important, growth of InN is faster in an inert carrier gas, like N_2 or He [12].

Since in both MOCVD and MBE In is found to act as a surfactant, it might also play this role in the HVPE growth of GaN. Because the growth temperature is high, no or very little In is expected to be incorporated into the layer. However, the positive effects of In on the quality of GaN grown by MOCVD and MBE could still be present for growth by HVPE. The In, which is assumed to be present at the surface, may change the growth kinetics and the roughness of the grown layers may decrease. This would be especially interesting for high growth rates in which roughening of the surface is more pronounced and thus poses a problem.

2. Experimental details

In our experiments, we used a home-built, horizontal HVPE reactor equipped with a rotating disc susceptor [15]. Pieces of a 2 in GaN/sapphire wafer are used as a template for HPVE growth. All templates used for the experiments described are cut from the same wafer, which is prepared by the gallium treatment step (GTS) method [16]. In general, MOCVD templates obtained with this method allow the growth of thick (300 μm) GaN layers without cracks by HVPE [17]. Growth experiments are performed at normal temperatures (1000–1100 °C); the carrier gas is either N_2 or H_2 (2800 sccm). The other gasses used are NH_3 (1000 sccm) mixed with N_2 (60 sccm) and HCl (50 sccm) mixed with N_2 (200 sccm). The boat used to place the gallium for growth has two compartments (see Fig. 1). The bottom of one of the compartments is always completely covered by Ga, the other one can be filled with In. The width of the boat is constant over the entire length (22.2 mm), the lengths of the compartments with In and Ga are 19.2 and 72.7 mm, respectively. This leads to a ratio between the In and Ga surface areas of 4:15. Before growth, the In is cleaned by etching in an aqueous HCl solution.

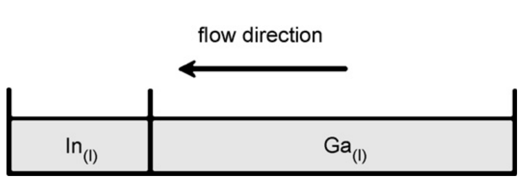

Fig. 1. Ga-boat with both In and Ga; the HCl gas first flows across the liquid Ga and then over the liquid In. The surface ratio of In and Ga is 4:15.

The growth species GaCl and InCl are formed in situ by passing HCl over the Ga-boat at 850 °C (see Fig. 1). The HCl first passes over the liquid Ga surface and then over the liquid In surface. Experiments were performed to see if the order of In and Ga had any influence on the growth. No effect on the growth rate was found. This shows that the amount of GaCl that is transported into the reactor does not depend on the order of In and Ga in the boat. The configuration of first passing the HCl over Ga and then over In is the easiest experimental option.

To investigate the effect of In with N_2 and H_2 as the carrier gas, four growth runs are performed: two without In as a reference, and two with In. The HCl flow through the boat is kept constant for all four experiments. The growth time is 1 h. An overview of the experiments is shown in Table 1.

After growth the surfaces of the samples are studied with differential interference contrast microscopy (DICM), two-beam interferometry [18,19] and atomic force microscopy (AFM; tapping mode).

3. Results and discussions

3.1. Growth rate

The growth rates for the different growth runs are reported in Table 1. The letters A, B, C, and D will be used to refer to the growth conditions. The highest growth rate is found for growth with N_2 as the carrier gas (sample A). When In is added to the growth system the growth rate decreases (sample B). A similar effect is seen for H_2 as the main carrier gas: again the growth rate decreases when In is added (samples C and D). The decrease in growth rate does not depend on whether the HCl flow first passes the Ga and then the In or the other way around. The same amount of GaCl, which is the species that determines the growth rate of GaN, is transported into the reactor for both cases. We have also seen that the HCl flow is not saturated with Ga after passing the boat, as the growth rate increases when the surface area of the Ga is increased. This means that InCl can be formed with the HCl that remains [10,14] and In is transported into the reactor. This also means that the same amount of GaCl enters the reactor irrespective of whether In is present or not. Apparently In influences the growth mechanism.

As both In and Ga are present in the gas phase, it can be assumed that both species will be present at the surface as well. It can be expected that the In atoms will only be incorporated into the bulk material at very low levels, because of the weaker In–N bond as compared to the Ga–N bond. However, the reduced growth rate in the experiments where In is added indicates its presence at the surface. There it will probably block positions where