



Effect of TMBi supply on low-temperature MOVPE growth behavior of GaN



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ABSTRACT

Undoped GaN and diluted GaN_{1-x}Bi_x alloys were grown on (0001) sapphire substrate by metal–organic vapor phase epitaxy (MOVPE) at 480 °C. By using in-situ laser reflectometry, it is found that the increase of TMBi flow rate leads to a reduction of the average value of reflectivity oscillations. Scanning electron microscopy (SEM) images gave a clear observation of the TMBi increasing amount effect on the surface morphology. The appearance of different structure (islands and columns) on GaN surface could be responsible to the reduction of the reflectivity oscillations average value. The energy dispersive X-ray (EDX) analysis showed that the observed structures were only composed of Bi compared to the flat GaN surface. Moreover, the surface morphology between islands and columns is improved when we increase the TMBi flow rate. This improvement is consistent with the decrease of root mean square (RMS) roughness, as measured by atomic force microscopy (AFM).

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1. Introduction

For many years, research has been conducted on the realization of optoelectronic devices with high quality such as light-emitting diodes (LEDs) and laser diodes (LDs). For this purpose, III–V nitride semiconductors like gallium nitride (GaN) have been the subject of numerous investigations. However, although the tuning of growth conditions like temperature, pressure and V/III ratio, GaN epilayers have a poor quality. This prevents the marketing of these devices. Surfactant assisted growth is a promising technique to improve epilayers quality. A surfactant element is typically characterized by a low solubility in the solid. As a result, it enhances the surface diffusion length and modifies the growth morphology of an epitaxial film without a high level of incorporation into the films. Therefore, several elements have been used. Additional of In during GaN growth without any noticeable In incorporation improves the GaN surface morphology [1,2]. Bi has been used previously as a surfactant for the growth of GaAs, In_xGa_{1-x}As [3], Ga_{Nx}As_{1-x} [4] and GaInP [5]. Recently, Bi has been used as a surfactant for the growth of GaN grown at 800 °C by molecular beam epitaxy (MBE) [6]. A similar Bi surfactant effect was observed for MOVPE GaN grown at 1050 °C [7]. More recently, it has been demonstrated that it is possible to grow the GaN_{1-x}Bi_x alloys by MBE at low temperature,

up to 80–90 °C [8,9]. A reduction of GaN band gap of about 200 meV/%Bi was obtained by increasing Bi content [9]. Therefore, GaN_{1-x}Bi_x based near infrared devices can be obtained.

GaN layers grown at low temperature by MOVPE on sapphire substrate exhibited a three-dimensional crystal growth such as large grains size. The use of Bi during low temperature GaN MOVPE-growth can improve the surface diffusion coefficient and result in a continuous film with a good surface morphology. No experimental information on the Bi effect has been reported yet on the low temperature GaN MOVPE growth.

Given the previous success of using Bi as surfactant for the growth of GaN or obtaining GaN_{1-x}Bi_x alloys, the growth of Bi-containing GaN at low temperature by MOVPE (LT-MOVPE) can provide further understanding of the Bi behavior in GaN. Therefore, in this study we report the effect of an additional TMBi flow rate on surface morphology of GaN grown by MOVPE at low temperature.

2. Experimental details

Undoped GaN and diluted GaN_{1-x}Bi_x alloys were grown on (0001) sapphire substrate in vertical MOVPE reactor operated at atmospheric pressure. The trimethylgallium (TMG), trimethylbismuth (TMBi) and ammonia (NH₃) were used as gallium (Ga), bismuth (Bi) and nitrogen (N) precursors, respectively. The carrier gas was a mixture of H₂ (2 slm) and N₂ (2 slm). The substrate was heated at 1080 °C under a mixture of H₂, N₂ and NH₃ during 10 min. Then, the temperature was decreased up to 480 °C and the TMG was introduced in order to grow undoped GaN sample. These samples were grown directly on nitridated sapphire substrate.

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Table 1
MOVPE growth parameters of diluted GaNBi alloys.

Samples	T_c (°C)	TMG		TMBi	
		(sccm)	($\mu\text{mol/min}$)	(sccm)	($\mu\text{mol/min}$)
A	480	5	19.8	0	0
B				3	1.7
C				5	2.8
D				8	4.5
E				10	5.6

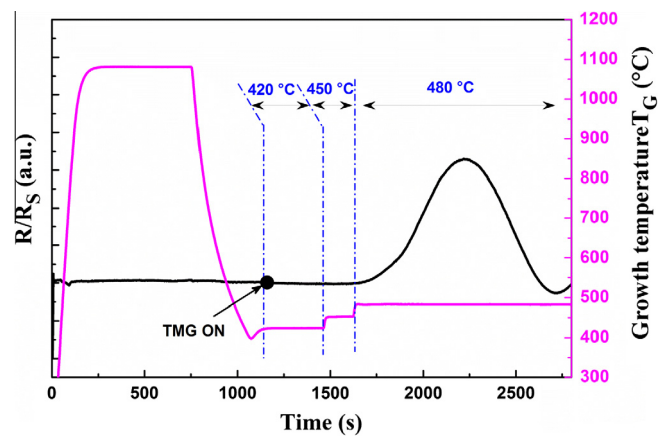


Fig. 1. Reflectivity signal and growth temperature recorded during the growth of GaN layer on sapphire substrate.

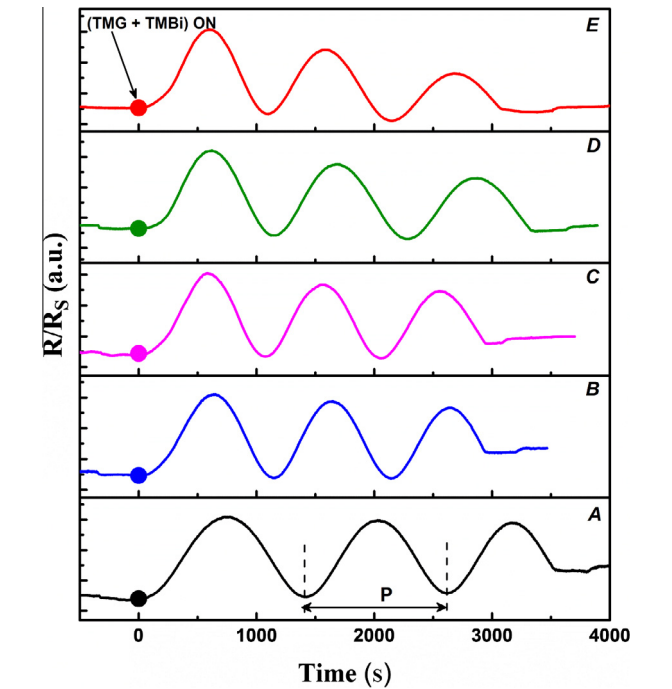


Fig. 2. Reflectivity signals recorded versus growth time during the deposition of diluted GaNBi alloys under TMBi flow rate of (A) 0 $\mu\text{mol/min}$, (B) 3 $\mu\text{mol/min}$, (C) 5 $\mu\text{mol/min}$, (D) 8 $\mu\text{mol/min}$ and (E) 10 $\mu\text{mol/min}$. Filled circle indicates the switching ON instant of TMBi and TMG into the reactor.

For the diluted GaNBi alloys, the TMG and NH_3 flow rates were maintained at 19.812 $\mu\text{mol/min}$ and 3 slm and an additional TMBi flow rate was introduced. The TMBi flow rate was varied from 0 to 5.575 $\mu\text{mol/min}$. The thickness of grown samples was kept at 0.38 μm . Details about the growth conditions of samples are summarized in Table 1.

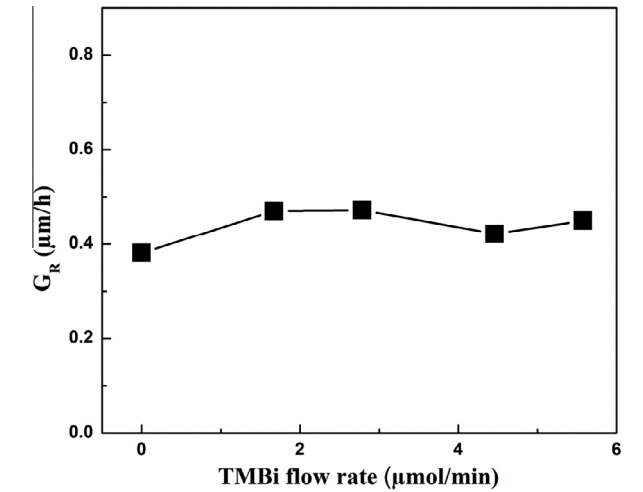


Fig. 3. Growth rate (G_R) of diluted GaNBi alloys as a function of TMBi flow rate.

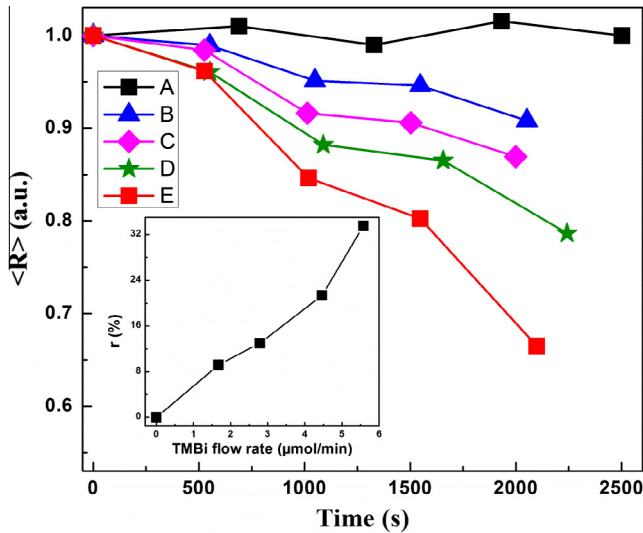


Fig. 4. Reflectivity oscillations average value ($\langle R \rangle$) of diluted GaNBi alloys as a function of growth time. The inset shows the relative reduction of $\langle R \rangle$ (r (%)) versus TMBi flow rate.

Growth was in-situ monitored by He–Ne laser reflectometry under normal incident light ($\lambda = 632.8 \text{ nm}$) [10,11]. The surface morphology was ex-situ examined by atomic force microscopy (AFM) operating in tapping mode and scanning electron microscopy (SEM). The chemical composition was analyzed by energy dispersive X-ray (EDX).

3. Results and discussion

The decreasing of the growth temperature results in an increase of Bi incorporation amount in GaN [8]. Consequently, the low growth temperature should be used to check Bi incorporation in GaN properties. Thus, we started this work by looking for the lowest growth temperature of GaN layer in MOVPE system. Fig. 1 shows the in-situ reflectivity signal recorded during the growth of undoped GaN layer by varying the growth temperature from 420 °C up to 480 °C. The reflectivity signal is normalized to the bare sapphire substrate as a reference. After a nitridation of sapphire substrate at 1080 °C, the temperature was decreased to 420 °C. In the following, a TMG flow rate is introduced into the reactor to achieve the GaN growth layer. At this temperature, as it can be seen from Fig. 1, the reflectivity signal stays constant, even at

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