The Influence of Substrate Orientation on Localized Nitrogen State in GaAsN films Grown on Vicinal GaAs (001) Substrates

Masaru Horikiri¹, Wen Ding¹, Yuki Yokoyama¹, Hidetoshi Suzuki¹, Tetsuo Ikari¹, Yoshio Ohshita², Masafumi Yamaguchi², and Atsuhiko Fukuyama¹

¹ Faculty of Engineering, Univ. of Miyazaki
1-1 Gakuen-Kibanadai-Nishi, Miyazaki 889-2192, Japan
Phone: +81-0985-58-7356 E-mail: ha11036@student.miyazaki-u.ac.jp
² Toyota Technological Institute
2-12-1, Hisakata, Tempaku, Nagoya 468-8511, Japan

Abstract

We investigated the influence of substrate orientation on the localized nitrogen state (E_N) in GaAsN films. The GaAsN films with nitrogen composition of about 0.6% were grown on vicinal GaAs (001) substrates by chemical beam epitaxy method, where substrate orientations were (001) tilted at 2 to 10° toward [010] direction. The energy levels of E_N were estimated from two split conduction subbands of E_+ and E_- measured by photoreflectance technique. E_N was found to decrease with increasing off angle. We considered that the decrease of E_N was caused by changing nitrogen incorporation.

1. Introduction

Dilute nitride semiconductor of InGaAsN has been expected as an absorbing layer material for ultra-high-efficiency four-junction solar cells such as InGaP/GaAs/InGaAsN/Ge structure [1]. This is because the band gap energy (E_q) of 1.0 eV and the same lattice constant as GaAs and Ge are achieved by controlling the composition of indium and nitrogen. In this case, indium can be used to compensate the nitrogen-induced reduction of the lattice parameter. It is known that E_q of GaAsN decreases drastically with increasing the nitrogen composition. This anomalous behavior of E_q has been understood in terms of a band anti-crossing (BAC) model [2]. In this model, an anti-crossing interaction between the localized nitrogen state (E_N) and the semiconductor matrix leads to a lowering of E_q . Therefore, the E_N is one of the most important parameters for deciding the band structure. So far, E_N have been reported to be a constant at 1.65 eV [2]. Recently, we found that E_N decreased with increasing nitrogen content. This indicated that E_N was modified by distribution of nitrogen atoms in the film, which was considered to be origin of low electron properties of GaAsN [3]. However, detailed relationships between nitrogen distribution and electrical properties are not understood yet. In our previous study, we reported that when the GaAsN films were grown on a vicinal GaAs substrate by using chemical beam epitaxy (CBE) method, those electrical properties were improved [4]. It is believed that this improvement is attributed to uniform distribution of nitrogen atoms in the film. Usage of vicinal substrate enhanced density of the surface step, which is the main incorporation site of nitrogen atoms during growth. Continuously, nitrogen atoms are incorporated uniformly by using the

vicinal substrates. According to the results, it is expected that E_N is also modified by vicinal GaAs substrates. However, the relationships between E_N and substrate orientation are not studied yet. In this study, we investigated the influence of substrate orientation of CBE-grown GaAsN films on the energy level of E_N by using the photoreflectance (PR) technique.

2. Experimental Procedures

Nondope GaAsN thin films were grown on vicinal GaAs (001) substrate by CBE method. The substrate orientations were (001) tilted at 2, 6, 8, and 10 degrees toward [010] direction. The films were labeled as *n*AB, where *n* indicates off angle of the substrates. The growth temperatures were fixed at 420°C. Detail growth conditions were listed in our previous paper [4]. All GaAsN films showed *p*-type conductions and their thicknesses were 1.0 µm. The nitrogen compositions of all samples were estimated to be about 0.6% by using the Xray diffraction method. The PR measurements were carried out using a standard setup at room temperature [5]. A modulated Ar⁺ laser (488 nm, 3.0 mW) was used as the excitation source to generate an electric field. The probe light for measuring a surface reflection was incident on the GaAsN surface at angle of 45°. The AC and DC components of the surface reflection were measured and the ratio of them $(\Delta R/R)$ was calculated. In the present study, since the reflection was also modified by surface morphology, surface roughness was evaluated by the atomic force microscope (AFM) technique. The measuring range of the AFM is limited to the surface of $5.0 \times 5.0 \ \mu m^2$.

3. Results and Discussion

Figure 1 shows $\Delta R/R$ spectra for all samples at room temperature. Three transition energies were observed. The E_{-} and E_{+} denote the transitions between valence band and the two split conduction subbands, respectively. They were caused by an anti-crossing interaction between E_{N} and the GaAs semiconductor matrix. In this case, E_{-} means E_{g} of GaAsN. The transition energy of $E_{-} + \Delta_{so}$ ($\Delta_{so} = 0.34$ eV) is that indicates the transition between the spin-orbit split-off valence band and E_{-} was also observed. As shown from figure, the signal intensities decreased with increasing off angle, and $E_{-} + \Delta_{so}$ and E_{+} were not observed in the 10AB film. From the AFM measurement, surface roughness of film was



Fig. $1\Delta R/R$ spectra of GaAsN films grown on vicinal GaAs substrates measured at room temperature.

constant that independent to the surface orientation. Thus, the decrease of signal intensity in $\Delta R/R$ was not caused by surface roughness but by quality of the film.

The transition energies of E_{-} and E_{+} were estimated by fitting the third derivative Aspnes' function to the $\Delta R/R$ spectrum [6]. Based on the BAC model, values of E_{-} and E_{+} are calculated by using the following expression,

$$E_{\pm} = \left(E_N + E_M \pm \left[E_N - E_M^2 + 4C_{NM}^2 x \right]^{0.5} \right) / 2 \quad (1)$$

where, x, E_M , and C_{NM} are nitrogen composition, the energy of the conduction band edge of unperturbed GaAsN, and the coupling parameter between E_N and E_M [2]. Thus, E_N can be calculated by using Eq. (1) as

$$E_{N} = E_{-} + E_{+} - E_{M} \tag{2}$$

Since E_M can be obtained by linear approximation of E_g of GaAs and GaN, E_N of each film can be calculated by using the measured transition energies of E_- and E_+ except the 10AB sample. Results were plotted in Fig. 2. Although E_N is reported to be a constant at 1.65 eV in the previously published data [2], calculated E_N decreased as increasing the substrate orientation. It is noted that the energy level of E_N depended on the distribution of nitrogen-induced localized states such as isolated nitrogen (N_x) and pair of nitrogen atoms (NN_i, where i = 1, 2, and so on) [3]. Moreover, it is reported that preferential nitrogen incorporation is related to step density [7]. Therefore, our experimental result suggested the variation of nitrogen distribution by changing nitrogen incorporation. As a result, it was suggested that the electron properties depended on nitrogen distribution.



Fig. 2 E_N in GaAsN films grown on vicinal GaAs substrates as a function of off angle.

3. Conclusions

We investigated that the influence of substrate orientation on E_N in GaAsN thin films grown on vicinal GaAs substrages. E_- and E_+ were observed at room temperature by photoreflectance measurements. Although the surface roughness was not change, the reflectivity decreased with increasing off angle of the substrate. In addition, E_N was found to decrease with increasing off angle. We considered that the change of E_N is caused by the change of distribution of nitrogen. These highly suggested that the electron properties of GaAsN films depend on the distribution of nitrogen in the film.

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