FABRICATION OF SELF-ASSEMBLING AlGaN QUANTUM DOT ON AlGaN SURFACES USING ANTI-SURFACTANT

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ABSTRACT

We report on the first artificial fabrication of self-assembling AlGaN quantum dots (QDs) on AlGaN surfaces using metal organic chemical vapor deposition (MOCVD). The AlGaN QDs are fabricated using a growth mode change from 2-dimensional step-flow growth to 3-dimensional island formation by modifying the AlGaN surface energy with Si anti-surfactant. The average lateral size and the thickness of fabricated AlGaN QDs, as determined by AFM, are approximately 20 nm and 6nm, respectively. The dot density was found to be controlled from 5×10^{10} cm^{-2} down to 2×10^{9} cm^{-2} by increasing the dose of Si anti-surfactant. We obtained the photoluminescence (PL) from AlGaN QDs embedded with $Al_{0.38}Ga_{0.62}N$ capping layers. The Al incorporation in AlGaN QDs was controllable within the range of 1-5 %.

INTRODUCTION

GaN and related nitrides are currently of great interest for the application to optical devices in the visible and ultraviolet (UV) energy range. The progress of blue laser diodes (LDs) or blue-green light-emitting diodes (LEDs) are extremely remarkable in recent years [1-3]. High-power and long-lifetime InGaN multi-quantum well lasers was already achieved[1].

AlGaN alloy is useful material for UV optical devices, because of wide bandgap direct transition emission between 3.4eV (GaN) and 6.2eV (AlN). The wide transition range of AlGaN covers the lasing wavelength range achieved by UV gas or solid state lasers, for example, XeCl(308nm) or KrF(248nm) excimer lasers, N_2(337nm), He-Cd(325nm) or SHG-Ar(257nm) lasers. UV semiconductor lasers are very attractive in comparison with gas or solid state lasers because of small size, long lifetime, high efficiency, low driving power and CW lasing operation. CW-UV lasers using AlGaN material in the near future will take the place of UV gas or solid state lasers.

For the realization of the UV semiconductor lasers, we should clear several technical barriers such as current injection through high Al content AlGaN crystals or efficient UV emission from AlGaN alloy. Especially, the realization of high optical gain in UV emission range of AlGaN alloy is most important for using it as active region of UV lasers. However, it is theoretically predicted that the reduction of the transparency carrier density is difficult in III-nitride material lasers because of a large effective mass [4] in comparison with GaAs or InP based materials.

In order to realize the high optical gain necessary in UV semiconductor lasers, the use of low dimensional quantum structures is quite useful. Optical gain enhancement using a exciton-related emission is theoretically predicted and expected even at room temperature, due to the large binding energy of exciton in GaN-related quantum dot structures [5]. It has been reported that the quantum efficiency of InGaN-based quantum well lasers is enhanced by the effect of localized excitons in nano-scale In segregated (In-rich) regions of the quantum well [6]. It is desirable to obtain a strong exciton-related emission from AlGaN quantum dot (QD) structures in order to achieve UV

lasers.

Recently, we have succeeded in the fabrication of self-assembling GaN and InGaN QD on AlGaN surfaces using metal organic chemical vapor deposition (MOCVD) and observed a strong photoluminescence (PL) emission from QD structures [7][8]. We also achieved optical pumping stimulated emission from GaN QDs sandwiched with AlGaN optical confinement layers[9]. The QDs were fabricated using a growth mode change from 2-dimensional step-flow growth to 3-dimensional island formation by modifying the surface energy balance of AlGaN with a Si anti-surfactant. Contrary to Stranski-Krastanov (SK) growth mode, the QD formation using this method is based on the surface energy balance. Then, this method is especially useful for the QD formation on the interfaces of small lattice mismatch system.

In this report, we demonstrate the first fabrication of self-assembling AlGaN QDs on AlGaN surfaces using MOCVD. The formation of AlGaN QDs on AlGaN surfaces is relatively difficult in comparison with the cases of GaN or InGaN QDs, because AlGaN easily forms a film on GaN or AlGaN surfaces due to large surface energy of AlGaN. The use of Si anti-surfactant was not enough to obtain a growth mode control to 3-dimensional nano-scale dot formation in high growth temperature. In addition to the use of anti-surfactant, we reduced the growth temperature of AlGaN QDs in order to control the migration of precursors on AlGaN surface, and as a result, we succeeded in the fabrication of nano-scale AlGaN QDs.

EXPERIMENTS AND DISCUSSIONS

The structures were grown, at 76 Torr on the Si-face of an on-axis 6H-SiC(0001) substrate, by a conventional horizontal-type MOVPE system. As precursors ammonia (NH_3), tetraethylsilane (TESi), trimethylaluminum (TMAl), and trimethylgallium (TMGa) were used with H_2 as carrier gas. N_2 gas was also independently supplied by a separate line and mixed with the H_2 just before the substrate susceptor. Typical gas flows were 2 standard liters per minute (SLM), 2 SLM, and 0.5 SLM for NH_3, H_2, and N_2, respectively. The molar fluxes of TMG and TMA of $Al_{0.38}Ga_{0.62}N$ growth for buffer and capping layers were 38 and 13 μmol/min, respectively. At this condition, the growth rate was approximately 2.5 μm/h. The molar fluxes of TMG and TMA of $Al_{0.05}Ga_{0.95}N$ growth for fabrication of QD structure were 7.2 and 0.47 μmol/min, respectively. The growth rate used for QD formation was approximately 0.4 μm/h. The substrate temperature during the growth was measured with a thermocouple located at the substrate susceptor.

The sample structure is shown in Fig.1. In order to achieve a surface suitable for growth of AlGaN QDs, first an approximately 400-nm-thick $Al_{0.38}Ga_{0.62}N$ buffer layer was deposited on a 6H-SiC substrate at 1140°C. The buffer layer was found to provide a step-flow grown surface as confirmed by atomic force microscopy (AFM). Prior to the $Al_xGa_{1-x}N$ dot growth, TESi was intentionally supplied for the deposition of silicon anti-surfactant modifying the surface properties at 1140°C. Then the sample was cooled to the $Al_xGa_{1-x}N$ dot growth temperature, i.e., around 900°C. The $Al_xGa_{1-x}N$ QDs were grown by a short supply of TMAl/TMGa using H_2 as carrier gas. The equivalent layer thickness, based on the growth rate, was determined to be 3 nm. This resulted in a three-dimensional nano-scale island growth which was not observed in the case without any silicon dose. Then an approximately 5-nm-thick $Al_{0.38}Ga_{0.62}N$ capping layer was grown on $Al_xGa_{1-x}N$ QDs.

In this experiment, we used relatively low growth temperature even for AlGaN QD formation in order to control the surface migration of precursors. So, we confirmed at first the atomically flat surface of AlGaN grown with low temperature. Figure 2 shows AFM views of the surface of $Al_{0.05}Ga_{0.95}N$ quantum film without using anti-surfactant grown at 900°C. A step-flow image is observed on the surface of AlGaN quantum film even in low growth temperature as seen in Fig. 2.

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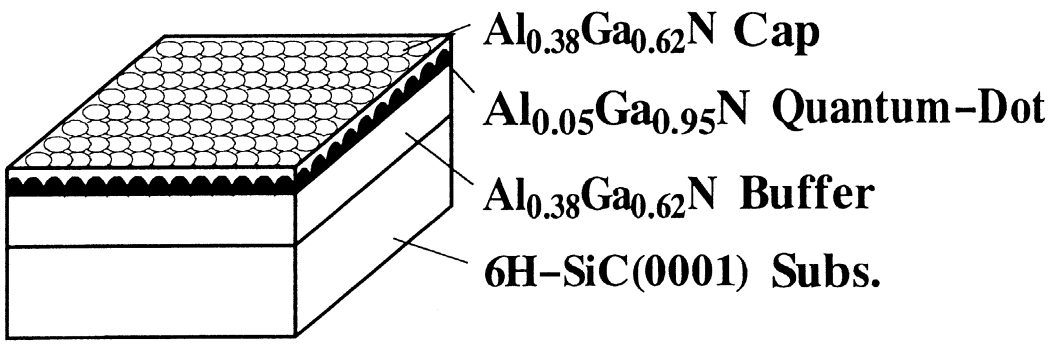

Fig.1. Schematic view of fabricated AlGaN quantum dot sample.

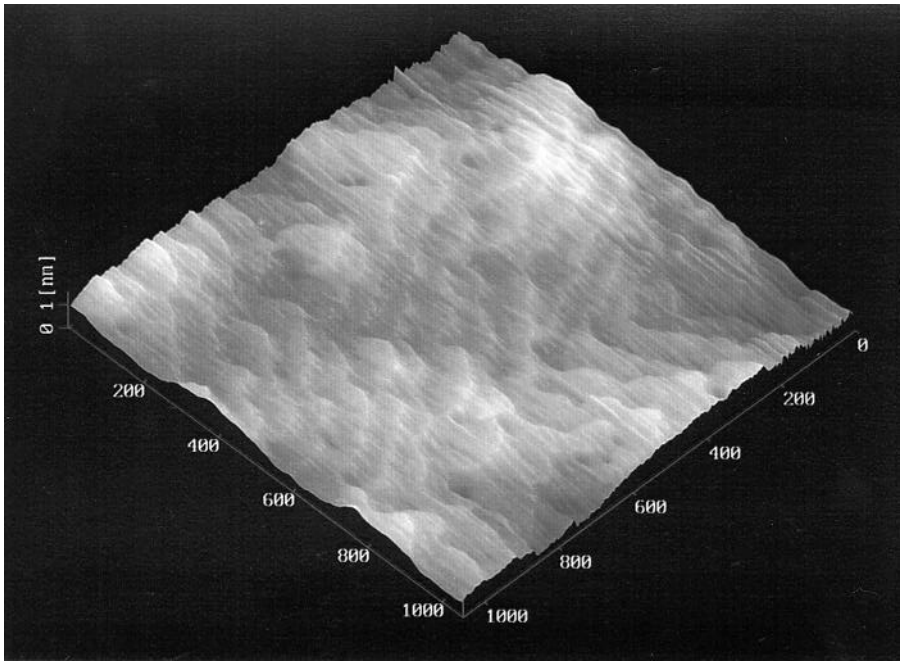

Fig.2. A Step-flow image of the surface of $Al_{0.05}Ga_{0.95}N$ quantum film grown without using anti-surfactant at 900°C observed by AFM.

Figures 3(a)-(d) show the AFM images just after the growth of $Al_{0.05}Ga_{0.95}N$ on Si deposited surfaces. The growth temperature Tg and the Si-dose amount are 1100°C and 0.04μmol, 1100°C and 0.2μmol, 900°C and 0.04μmol, and 900°C and 0.2μmol for Fig. 3(a), (b), (c) and (d), respectively. 3-dimensional growth is seen on the samples using anti-surfactant. For high growth temperature of 1100°C, we could not obtain nano-scale dot structures as shown in Fig. 3(a) and (b). We can see an network-like morphology on the grown surface for small amount of Si-dose of 0.04μmol which is enough large amount for the fabrication of GaN or InGaN nano-scale QDs. We obtained large dot structures when increasing the Si-dose amount by 5 times as seen in Fig. 3(b). In this case, the typical lateral size of the dot was 100-200nm. Therefore, the surface energy control by using Si anti-surfactant was not enough to obtain a growth mode change to 3-dimensional nano-scale QD formation for AlGaN.

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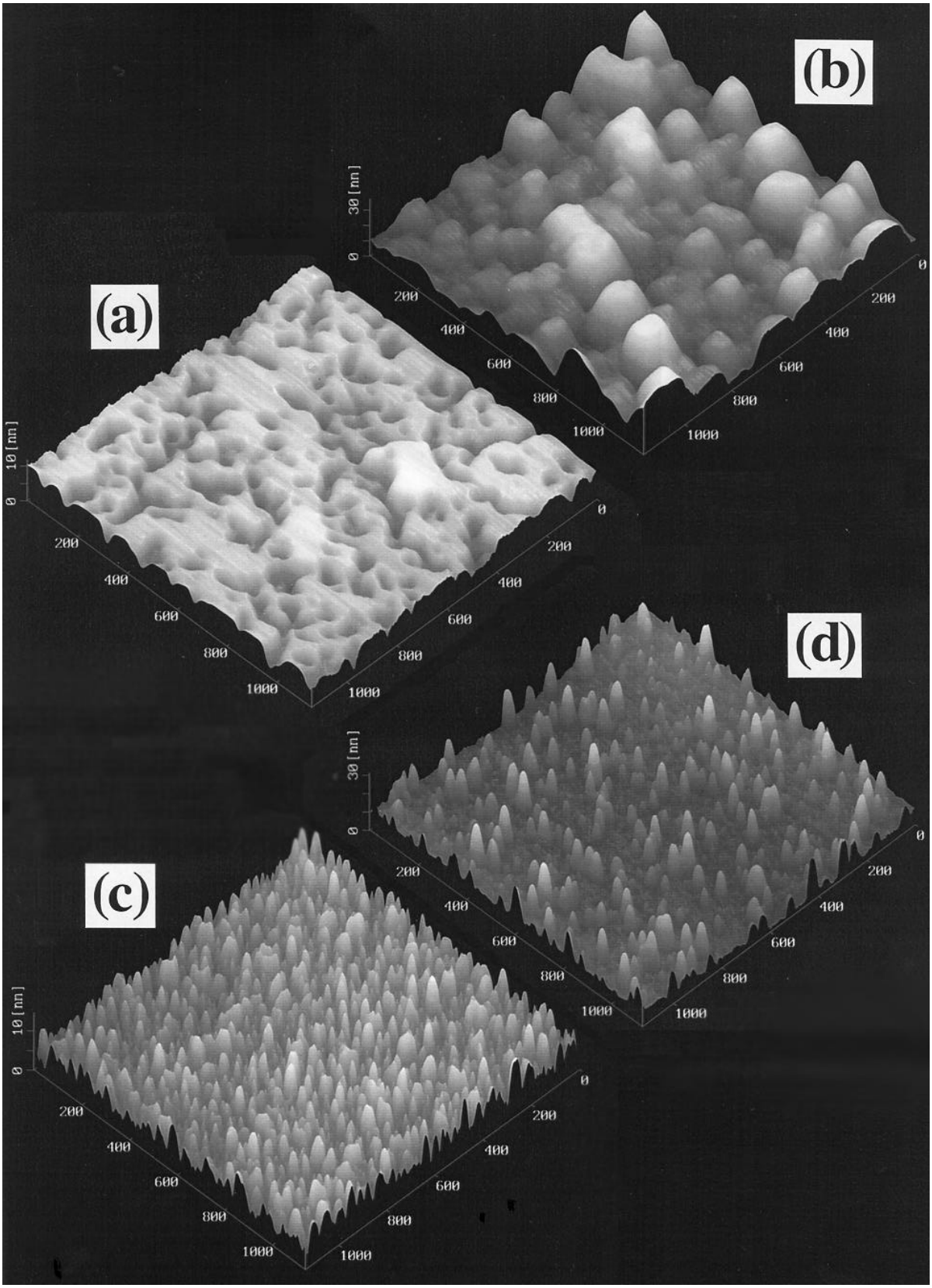

Fig. 3(a)-(d). AFM images just after the growth of $Al_{0.05}Ga_{0.95}N$ on Si deposited surfaces. The growth temperature Tg and the Si-dose amount are (a)1100°C and 0.04μmol, (b)1100°C and 0.2μmol, (c)900°C and 0.04μmol, and (d)900°C and 0.2μmol.

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On the other hand, the AlGaN dot was controlled to nano-scale structure by reducing the QD growth temperature to 900°C as seen in Fig. 3(c) and (d). The average lateral size and the height of fabricated AlGaN dots grown at 900°C were estimated to be approximately 20 nm and 6 nm, respectively, by AFM views. The dot density is found to be controlled from 5×10^{10} cm^{-2} down to 2×10^{9} cm^{-2} by increasing the dose of Si anti-surfactant from 0.04μmol to 0.2μmol.

We are investigating the GaN QD formation mechanism on AlGaN surfaces in detail [10]. However, the detailed formation mechanism is still very difficult to observe. It was found from our experiment that very small amount of Si atom deposited on the steps of AlGaN (several % of mono-layer) inhibit the step-flow growth of GaN, because the Ga atom is hard to be adsorbed around Si atom. The disordering process of step-flow growth makes a state of chaotic wandering and may stimulate the transition to 3-dimensional GaN growth. We guess that Al atom is much easily to be adsorbed around Si in comparison with Ga atom, therefore, the transition to 3-dimensional growth mode is hard to occur. The successful formation of nano-scale AlGaN QDs may be due to the enhanced effect of step-flow inhibition with the reduction of Al migration on the surface.

We obtained the photoluminescence from AlGaN QDs embedded with $Al_{0.38}Ga_{0.62}N$ layers. Figure 4 shows 77K PL spectra from AlGaN QDs with capping layers fabricated at 900°C with the Si dose of 0.04μmol for various TMAl flow rate. The emission intensity was the same level as those of GaN QDs fabricated with the same growth condition. The blue shift of QD emission due to the increase of Al incorporation was observed. Al molar fraction in QDs are estimated to be 0.01, 0.03 and 0.05 for the TMAl flow rate of 0.19, 0.38 and 0.47μmol/min, respectively, taking into account the quantum confinement level shift.

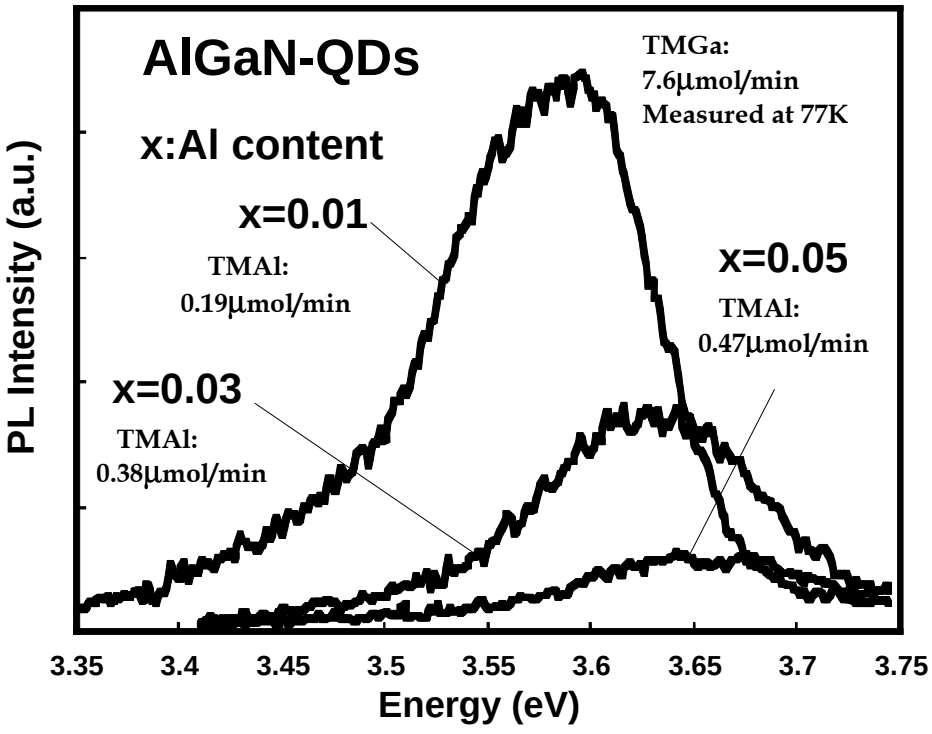

Fig. 4. 77K PL spectra from AlGaN QDs with capping layers fabricated at 900°C for various TMAl flow rate.

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At this moment, the half width of the AlGaN QD emission is still large which may be mainly caused by the size fluctuation of QDs. Also the emission intensity is week for high Al content QDs. The emission intensity and size fluctuation of QD may be much improved even for high Al content QDs by increasing the growth temperature of QDs and by optimizing the growth condition of buffer and capping layer. We are now trying high temperature AlGaN QD growth by controlling the surface migration of precursors with the use of more heavy carrier gas.

CONCLUSION

We demonstrated the first artificial fabrication of self-assembling AlGaN QDs on AlGaN surfaces using low pressure MOCVD system. The AlGaN QDs were fabricated using a growth mode change from 2-dimensional step-flow growth to 3-dimensional island formation by modifying the surface energy balance of AlGaN with Si anti-surfactant. The average lateral size and the thickness of the AlGaN QDs, as determined by AFM, were approximately 20 nm and 6nm, respectively. The dot density was found to be controlled from 5×10^{10} cm^{-2} down to 2×10^{9} cm^{-2} by increasing the dose of anti-surfactant. We obtained the photoluminescence from AlGaN QDs embedded with $Al_{0.38}Ga_{0.62}N$ layers. The Al content of AlGaN QDs was controllable within the range of 1-5 %.

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