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Invited

Fabrication of GaN Quantum Dots and Their Optical Properties

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1. Introduction

In recent years, GaN and other group III nitrides have been extensively investigated, largely in order to achieve high quality short wavelength light emitting and laser diodes (LED/LD). Laser action from InGaN multi-quantum well structures was recently achieved by Nakamura et al. [1]. Among many problems needing to be solved for the future applications of these devices, obtaining a low threshold current density is essential. By reducing the dimension of the active layer structure, i.e. to that of a dot, if the size of structures is at least comparable to the effective Bohr radius of an exciton, then this value may be improved due to increased binding energies of excitons. Furthermore, a variety of physical phenomena due to many body effects of excitons is of great interest, for example, a biexcitonic transition [2] from the nanoscale GaN crystal is particularly interesting as a possible method to achieve new LD devices, quantum dot lasers.

Quantum dot research has been very active in group III-V materials. Indium or phosphine based III-V compound quantum dots were successfully fabricated by the Stranski-Krastanow (S-K) growth technique, where quantum dots can *spontaneously* form on a lattice mismatched substrate. GaN quantum dots are, however, essentially difficult to fabricate on $Al_xGa_{1-x}N$ surfaces, which are indispensable for carrier and optical confinements for shortwavelength quantum dot laser diodes, by the S-K growth mode. In this system, two dimensional growth or step flow inevitably occurs because of two fundamental reasons; a small lattice mismatch between a film and substrate and a favorable surface free energy balance for two dimensional growth [3]. In this paper, we report a new approach for GaN quantum dot formation on AlGaN surfaces, where no S-K growth occurs, by artificial modification of the surface free energy of the AlGaN surface by means of Si atoms.

2. Experimental

Initially, the multilayer structure consisting of a ~0.4 μm thick $Al_{0.12}Ga_{0.88}N$ barrier layer (or compositionally graded $Al_xGa_{1-x}N$ (x = 0.12 - 0.2) layer) and a ~0.6 μm thick $Al_{0.20}Ga_{0.80}N$ cladding layer was deposited at 1080 °C on the Si face of an on-axis 6H-SiC(0001) surface using a conventional horizontal-type metalorganic chemical vapor deposition (MOCVD) system. After depositing the $Al_xGa_{1-x}N$ multilayer structure, less than one monolayer of tetraethylsilane ($Si(C_2H_5)_4$: TESi) with the H_2 carrier gas was supplied on the AlGaN barrier layer surface, followed by a short pulse supply of TMG and NH_3 gases for the growth of GaN. For comparison, samples without a TESi dose step, resulting in a single quantum well, were also prepared. Surface morphologies in each sample were investigated by an atomic force microscope (AFM). Another set of samples which were capped with a thin layer of AlN were prepared to study optical properties by photoluminescence (PL) spectroscopy.

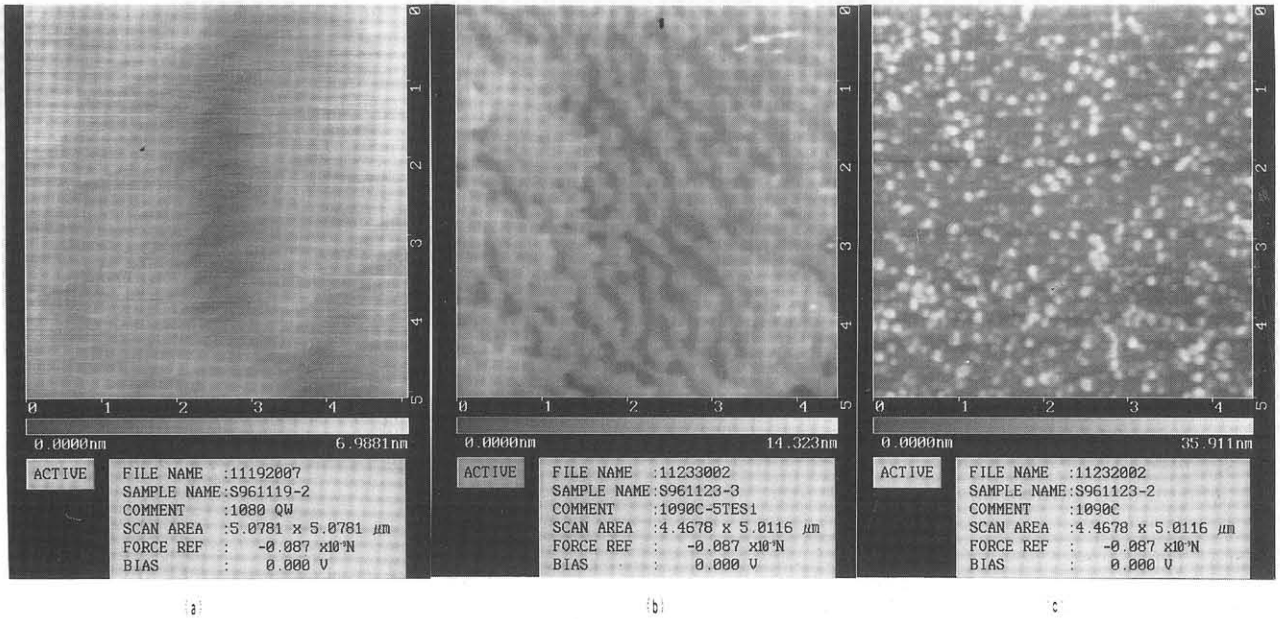

Fig. 1 AFM micrographs of GaN surfaces under several TESi dose rates.

3. Results

Growth mode

The effect of the total TESi dose on the resulting GaN surface morphologies is clearly shown in the AFM micrographs in Fig. 1 (a), (b), and (c). The total TESi dose in each sample was 0, 2.2 x 10^{-8} mol, and 2.2 x 10^{-7} mol in (a), (b), and (c), respectively. Here, the total dose simply represents the TESi gas feeding rate into the reactor. The net number of Si atoms/molecules arriving at the surface can be several orders less than this value, taking into account the areal ratio (r) of the substrate to the reactor and the reactive sticking coefficient (η) of the TESi gas. Assuming r = 0.01 and η = 0.01, the total number of Si species in (b) arriving at an AlGaN unit area (100 nm x 100 nm), which contains one 100 nm length of a step based on the AFM measurement, is calculated to be ~5 x 10^2. This value is in the same order of the number of atoms at the step site. The net number of Si atoms/molecules arriving at the surface can be several orders less than this value. Without introducing TESi (0 nmol) a step flow feature of GaN on the AlGaN surface is observed in Fig. 1 (a), where atomic steps whose heights are in average double bilayers (~0.5 nm) as determined by line scan measurements across the steps. This feature resembles the surface morphology of AlGaN barrier layer, indicative of step flow growth in this system. When the TESi dose was increased to 22 nmol, the GaN surface appears to experience certain morphological changes as shown in Fig. 1 (b). Fairly large sizes of GaN islands elongated along steps are observed. The growth mode transition from step flow to islanding seems to be controlled by the TESi dose. In another words, step flow growth is inhibited by adding Si atoms on the AlGaN stepped surface. A further increase of the TESi dose to 220 nmol introduces more drastic change of the GaN surface morphology, as indicated in Fig. 1 (c). Clearly isolated small islands, quantum dots, are formed at this stage, probably as a result of an enhanced inhibition of step flow growth and a decreased surface free energy. As the TESi dose was further increased, the dot density was decreased.

Optical properties

The optical property of the AlN capped GaN quantum dots at different temperatures was investigated by PL using a He-Cd laser (325 nm) as the excitation source. Figure 2 shows a typical PL spectra of the GaN quantum dots. The dot size of this sample was determined to be ~7 nm in height and <~20 nm in width using AFM. A peak, visible at 3.500 eV exhibits a blue shift of 18 meV based on the free exciton energy of 3.48 eV for an unstrained GaN film. The blue shift is probably due to both strain and quantum confinement effects. The peak shift corresponding to dot size change observed (not shown) especially supports the latter effect. A peak at higher energy is beleived to be due to defects in the AlGaN buffer layers. Moreover, a clear temperature dependence of the exciton peak energy is observed. It is noted that the exciton peak from GaN dots was visible even at 320K. Detailed discussions will be given in conjunction with quantuization energy calculations. PL experiments at higher excitation intensities using an N_2 laser (337 nm) were also performed on a sample having a multi-layer structure containing optical and carrier confinement AlGaN layers. A clear stimulated emission from the sample edge was observed [4]. This results demonstrate the potential of GaN quantum dots for use in LD applications.

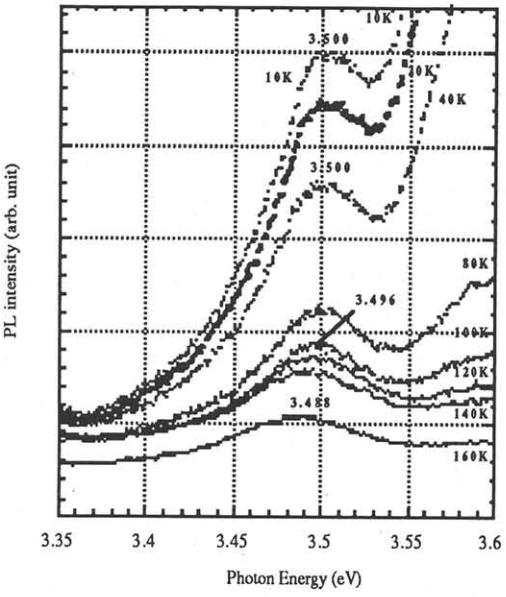

Fig. 2 PL spectrum from GaN quantum dots grown on AlGaN surface.

5. Conclusions

In conclusion, GaN quantum dots have been fabricated on AlGaN cladding surfaces using TESi, inhibiting the generally observed step flow growth in the GaN/AlGaN closely lattice matched systems. The change in growth mode was qualitatively discussed in terms of kinetic and energetic consideration, using the models of step flow inhibition and surface free energy balance, respectively. An implication for quantum dot fabrication in closely lattice matched (non S-K growth) systems was demonstrated. The optical property of GaN quantum dots clearly indicated the quantum confinement effect.

References

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