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Growth and Investigation of GaN/AlN Quantum Dots

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ABSTRACT

We have fabricated GaN quantum dots (QDs) in AlN confined layer structures by molecular beam epitaxy. The size distribution and density of the QDs have been estimated from an atomic force microscopy study. Very high quantum efficiency of photoluminescence (PL) has been obtained in some samples with QDs. Compared to the GaN bulk samples, it increased by orders of magnitude. In some samples the quantum size effect dominated, resulting in the blue-shift of the QD related PL peak, whereas in the samples with larger dots a red-shift up to 0.8 eV has been observed, which is related to strong polarization effects. We have observed a blue-shift of the PL peak with excitation intensity in the samples with large dots due to screening effect. The temperature-induced quenching of PL occurs at higher temperatures compared to bulk GaN due to the confinement of nonequilibrium carriers in the QDs. An excited state has been observed in some samples.

INTRODUCTION

Quantum dots (QDs) in nitride based semiconductors are expected to improve characteristics of the visible-to-UV optical emitters which are currently produced by employing two- and three-dimensional growth. The idea is that the layer or layers of quantum dots will decouple the active layers from the substrate or buffer layer and, thus reduce the density of extended and point defects. This application of QDs is novel. In the same vein, Gérard et al [1] applied dots for reducing the degradation of internal quantum yield. They pointed out that once the carriers are captured by QDs, they become strongly localized and their migration toward nonradiative recombination centers is made more difficult. In the absence of a surface topology driven process, the dots nucleate on dislocations that propagate to the surface of the layer. The key to high quality is then to obtain a dot density that is substantially larger than the dislocation density. The goal then should be to reduce the dislocation density below 10^8 cm^{-2} and increase the dot density above 10^{11} cm^{-2}. This would simply mean that less than one out of every 1000 dots would contain extended defects. Growth of GaN self-assembled QDs on AlGaN, with the aid a sub- to monolayer Si layer, which are then covered by AlGaN has been first reported by Tanaka et al. [2]. Other approaches have been reported by Widmann et al. [3,4] and Damilano et al. [5] who used AlN wetting layers which provide a larger lattice mismatch to GaN than AlGaN, and in turn provides the impetus for a 3-D growth. In addition, the surface topology of AlN is smoother than that of AlGaN, which removes the surface features from being the nucleation sites for dots. Dots have been demonstrated on 6H-SiC [2,6] and sapphire (0001) [3-5]. The growth was undertaken using metalorganic vapor phase epitaxy (MOVPE) [2,6] as well as molecular beam epitaxy (MBE) [3-5]. Depending on the size of the GaN dots, the blue-shifted emission [2-4,6] and red-shifted emission [4,5] have been observed. By changing the size of the quantum dots, one can in fact tune the color of the emission [5].

GROWTH AND EXPERIMENTAL DETAILS

GaN dots discussed here were grown on [111] Si or on c-plane of sapphire substrates by reactive MBE. Following a monolayer deposition of Al at a somewhat lower temperature, and nitridation of that Al layer, a nominally 0.3 μm thick AlN layer at 900 ^{o}C and a 1-2 μm thick GaN layer were grown on Si. More standard approaches were employed for growth on sapphire substrates. The GaN buffer layer was followed by the deposition of a sufficiently thick AlN layer, which is presumed to be relaxed. In a set of samples, the buffer layer was a 10 μm thick GaN grown by hydride vapor phase epitaxy (HVPE).

Quantum dots were formed by growing a GaN layer having a thickness, which allows it to maintain its coherency with the AlN lattice in a fashion similar to that reported in reference 5. A short thermal-anneal-process causes the dots to form. The initial thickness and the length of the annealing cycle determine the particular size of the dots. Some 20 to 30 bilayers of GaN/AlN have been grown on top of the sample with interruption providing the GaN dot formation.

For the optical study of QDs, the photoluminescence (PL) was excited with a continuous-wave He-Cd laser (photon energy 3.81 eV) or with the third harmonic of a "Tsunami" Ti-sapphire laser (photon energy 5.06 eV) and analyzed with a SPEX grating monochromator with a Hamamatsu photomultiplier tube (PMT) R955. The temperature of our samples was varied from 10 to 300 K using a closed-cycle cryostat.

RESULTS AND DISCUSSION

Figure 1a shows an AFM image of a single layer of GaN QDs grown on AlN layer by MBE. The AFM image of the sample with 20 bilayers of GaN/AlN is shown in figure 1b.

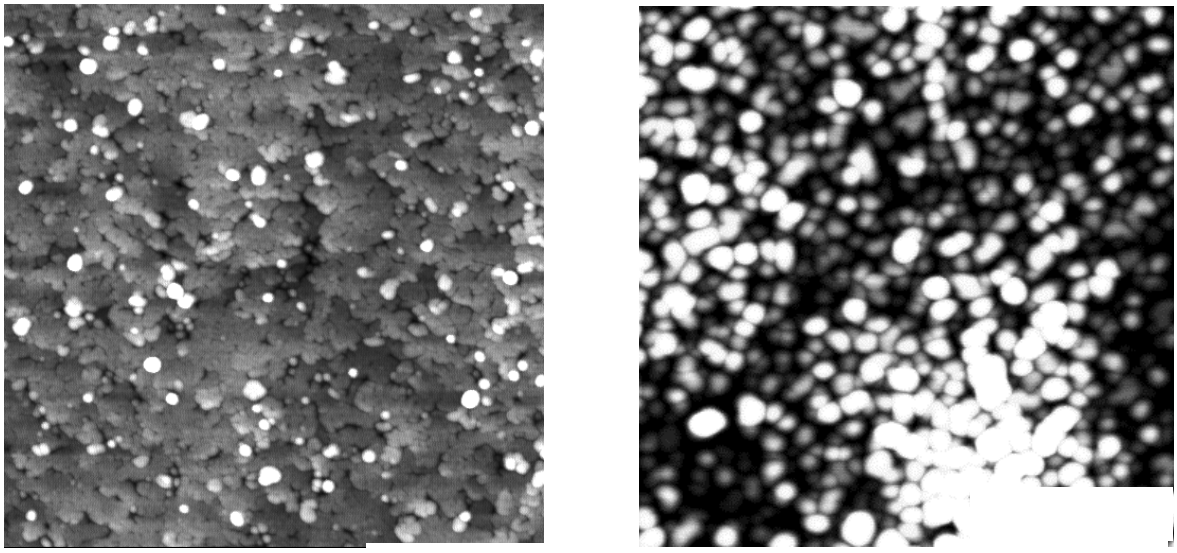

Figure 1. *(a) AFM image of single layer GaN QDs grown on AlN layer. The height of the dots is about 6 nm. The vertical scale varies between 0 and 12 nm. (b) AFM image taken on multi-layer GaN quantum dots grown on AlN over a ~1μm thick GaN buffer layer. The lateral size of the dots is roughly 40-90nm. The height of the dots is about or less than 10 nm. Note the high density of dots.*

In the PL spectrum of samples with GaN/AlN QDs, we observed bands, which were red-shifted or blue-shifted relative to the near-band-edge emission (at about 3.47 eV at 10 K), see figure 2.

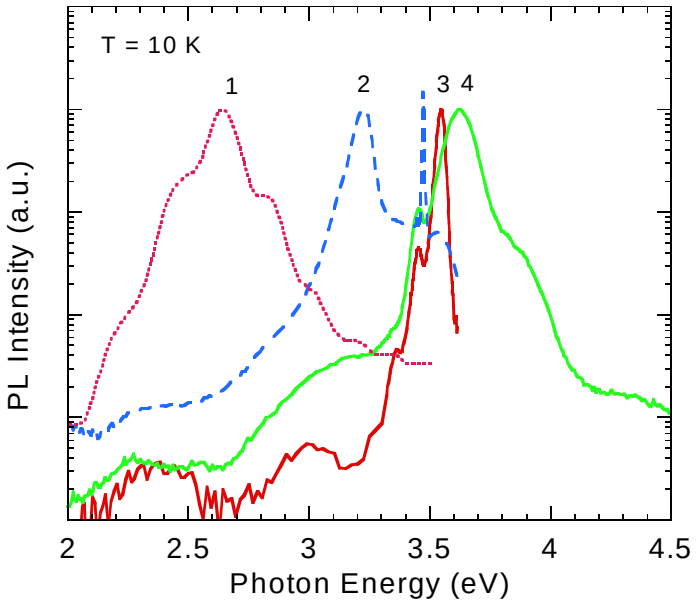

Figure 2. *PL spectrum of samples constituting the GaN/AlN QDs grown on Si (1) and sapphire (2-4) substrates with GaN buffer layers grown by MBE (1,4) and HVPE (2,3). The intensity is normalized at maximum of the QD-related maximum. The peak at about 3.47 eV is related to exciton emission in the GaN buffer layer. Note the log scale of the PL intensity.*

The bands were attributed to the QD structure since they appeared only in the samples with such structure and exhibited high PL intensities. The quantum efficiency of the PL from the samples with QDs was always much higher than that from the reference bulk GaN samples. The internal quantum efficiency amounted to 50 % in some samples on GaN grown by the HVPE method. The high efficiency is ascribed to strong localization of the carriers in QDs, which makes their migration toward nonradiative recombination centers difficult.

The red-shift of the QD emission relative to the excitonic emission in GaN is attributed to polarization effects, whereas the blue-shift is caused by the confinement effect. The red-shift increases with increasing height of the dots, while the blue-shift evidences small size of the dots. A competition between these two effects results in different positions of the QD peak in the PL spectrum (figure 2). Note that the red-shift due to the polarization effect at the GaN/AlN interface is very large, which is consistent with calculations and experimental results of Widman et al. [4]. With increasing excitation intensity, the PL from the QD region is blue-shifted. The shift was larger in samples with initially red-shifted position of the PL. We attribute the blue-shift of the PL bands with excitation intensity to screening. Different values of the shift in different samples reflect different lateral size and shape of the dots.

Another interesting effect has been observed in some samples at high excitation density. While the main peak attributed to QDs increased nearly linearly with excitation intensity, an additional peak appeared, whose intensity increased superlinearly (figures 3,4). We attribute this blue-shifted peak to the excited state of the QDs. In the sample with a red-shifted PL, the

distance between the ground and excited states is estimated as ~ 0.12 eV (figure 3). In the sample with the blue-shifted PL, it is about 0.27 eV (figure 4).

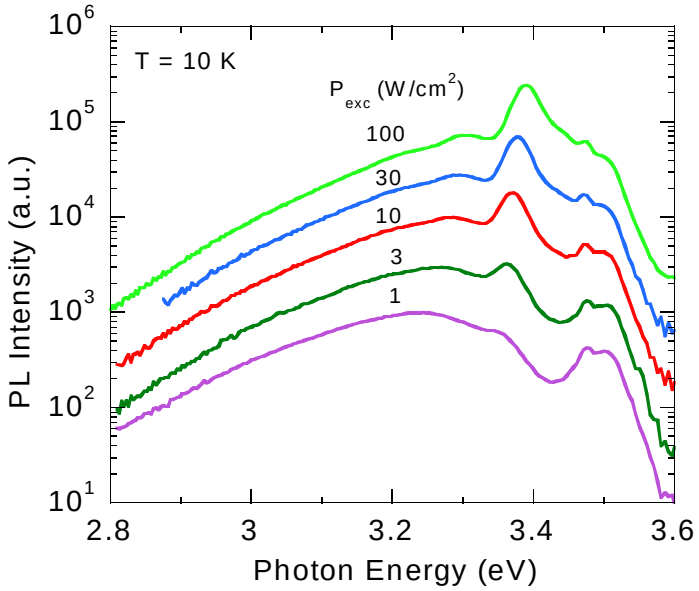

Figure 3. *Effect of excitation intensity on the PL spectrum of the sample with QD structure on top of a 10 μm thick GaN buffer layer grown by HVPE. A broad band with a maximum at about 3.2 eV is related to the QD structure. Another band with a maximum at about 3.35-3.40 eV appears only at high excitation intensities. It is related to the excited state of the QDs. A peak at about 3.47 eV emerges from the buffer GaN layer. Both 3.2 and 3.35 eV bands which are blue-shift with excitation intensity are due to screening.*

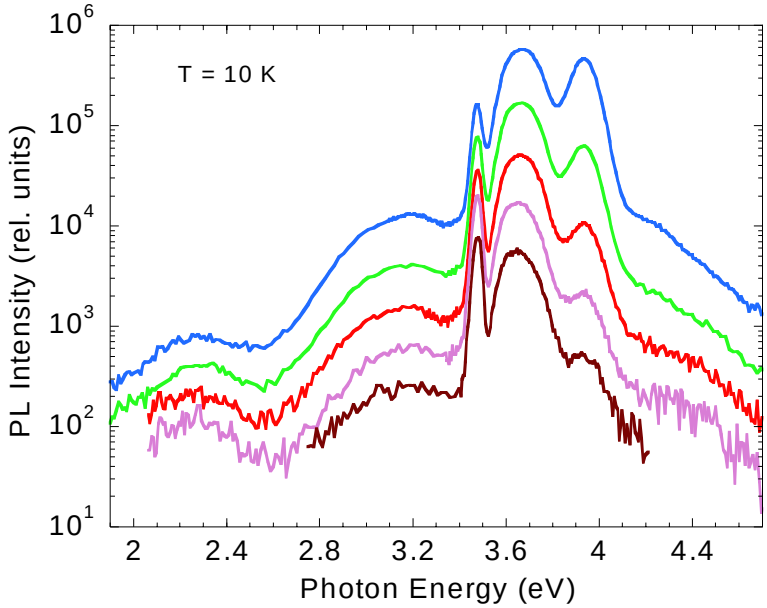

Figure 4. *Effect of excitation intensity on the PL spectrum of a sample with QD structure on top of 1 μm thick GaN buffer layer grown by MBE. The peaks at about 3.65 and 3.93 eV are attributed to the ground and excited states of GaN QDs. The peak at about 3.47 eV is related to excitonic transition in the GaN buffer layer.*

We studied the variation of PL intensity with temperature as well. The intensity of the QD emission remained almost unchanged up to about 100 K, then the thermal quenching took place with an activation energy of the order of 200 meV (figure 5). Interestingly, the temperature dependence was quite similar for the red- and blue-shifted emission from two samples with QDs, which confirms that the studied PL bands are related not to radiative defects in bulk GaN, but to exciton emission in the QD structure. The reduced temperature dependence compared to the bulk exciton emission is related to localization of the carriers inside the dots.

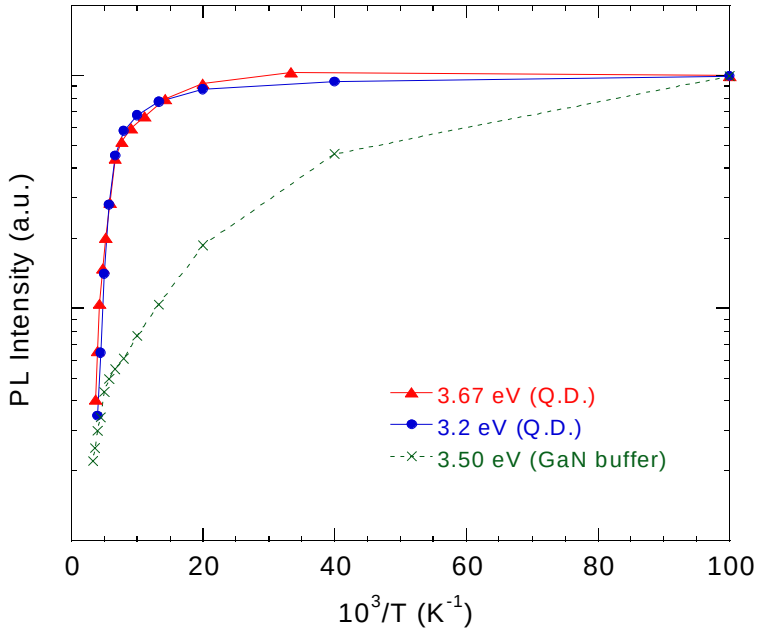

Figure 5. *Temperature dependence of the intensity of PL caused by exciton recombination in the GaN buffer layer and in the QD layer. The dependencies of two QD samples are shown to demonstrate that both the red- and blue-shifted QD bands have nearly the same temperature dependence.*

CONCLUSIONS

GaN quantum dots have been grown with reactive molecular beam epitaxy on Si and sapphire substrates. AFM images indicate a high density of well-defined dots with a large lateral size. PL measurements demonstrated high quantum efficiency of emission from the quantum dot structure. Different positions of the QD-related peak, from ~ 2.6 to 3.7 eV in different samples, resulted from the competition between the polarization and confinement effects. An excited state of the QDs in the PL spectrum was observed at high excitation intensities. A high quantum efficiency, along with the reduced temperature dependence of the QD PL intensity, evidence improved quality of the QD structure compared to bulk GaN.

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