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Original method of GaN and InGaN quantum dots formation on (0001)AlN surface by ammonia molecular beam epitaxy

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Abstract. We report original method of formation Ga(In)N/AlN quantum dots with low density by ammonia MBE on the (0001)AlN surface by using a decomposition process of Ga(In)N thin layer. Low density of quantum dots have been obtained in the range 10^7-10^9 cm^{-2}. Single quantum dots photoluminescence lines corresponding to exciton and biexciton transitions were observed in micro-photoluminescence spectra.

1. Introduction

Formation of the wide band gap semiconductor GaN and InGaN quantum dots (QDs) in AlN matrices are important for fabrication of modern quantum devices such as single electron transistors, single and entangled photons sources, UV light emitting diodes, lasers and detectors, and etc. The GaN and InGaN QDs formation at the (0001)AlN surface usually implemented by using of the Stranski-Krastanov growth mode [1], or by using the droplet epitaxy technique, where at first Ga(In) droplets are deposited onto the surface, and then droplets are transformed into GaN (InGaN) 3D islands as result of the treatment in active nitrogen flux (like NH_3) at temperatures about 700 °C [2]. Usually density of QDs ensemble is quite high as 10^{10}-10^{11} cm^{-2} [1,3]. But for fabrication of single electron transistors or single photons sources the density of QDs of 10^8 cm^{-2} is required.

In the present work we propose an original method for the low density GaN (InGaN) QDs formation by using an Ga(In) wetting layer on the (0001)AlN surface [4] to form controllable thin two dimensional (2D) GaN layer and subsequent decomposition of this layer.

2. Experimental

Samples were grown by molecular-beam epitaxy (MBE) on 2 inch (0001)-oriented sapphire substrates in a Riber CBE-32 machine with ammonia as a source of active nitrogen. Formation process of the quantum dots were studied *in situ* by high energy electron diffraction (RHEED). The obtained structures with QDs were investigated by micro-photoluminescence (micro-PL). Photoluminescence was excited by continuous-wave HeCd laser with photon energy of 3.81 eV. The laser beam was focused in the spot with diameter of 1 μm on the sample by micro-objective. The PL was analyzed by

Acton SP-750i spectrometer with nitrogen-cooled CCD detector. A helium cryostat was used to control of the sample temperature.

3. Results and discussion

We have studied a two dimensional to three dimensional (2D-3D) growth mode transition during epitaxial growth of GaN (InGaN) layers with rate of 0.25 monolayers per second (ML/s) under ammonia flux (200 sccm) in wide temperature range (400-1000 °C) in order to determine suitable conditions for preparing of low density quantum dots ensemble. When the growth of GaN initiated, first the 2D growth mode was observed, that is evidenced by streaky RHEED pattern of the growing surface. Then the 3D transmission spots were appeared and it was found that the time interval from the growth initiation to transition point strongly depends on the surface temperature. The 3D GaN spot intensity behavior is shown in Figure 1. It should be noted that the 2D GaN growth without 2D-3D transition was observed at temperatures higher 750 °C. At temperatures higher ~870 °C the GaN growth rate drops because of decomposition of the GaN layer.

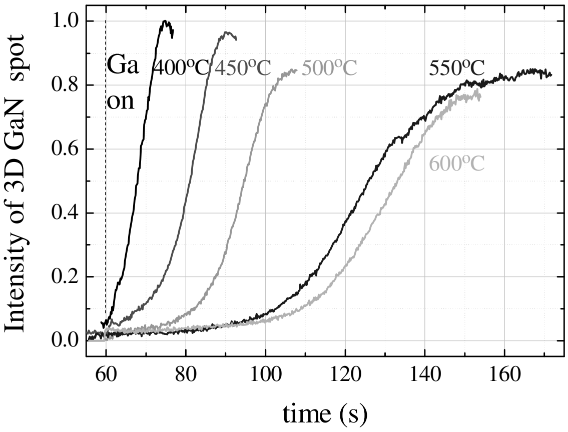

Figure 1. GaN 3D islands formation on (0001)AlN during epitaxial growth at different temperatures.

It is clear from the Figure 1 that the kinetic curves of 3D GaN islands (or GaN QDs) formation have a S-like character and the rate of this process decreases with increasing of temperature. It is evidence that the 3D GaN islands formation process is connected with a surface phase transition. The order parameter of the phase transition can be defined as densities difference between high density and low density phases of GaN QDs ensemble on the surface in close analogy with the van der Waals type of transitions from gas to liquid. However, this process is not fit to our goals because it results in high density of GaN QDs 10^9-10^{11} cm^{-2}.

We have developed an original method of GaN QDs formation. It is known that thermal decomposition of GaN occurs at temperatures higher 900 °C. The GaN decomposition is the multi-step process with appearing on the surface of GaN 3D islands [5].

Our QDs formation process consists of following stages. Three monolayers of Ga (or In+Ga) was deposited onto clean AlN surface at temperatures 500-600 °C. The RHEED pattern is quenched as a result of liquid Ga(In) layer formation on the surface. Then the surface was treated at the same temperature under ammonia flux (10 sccm) during of 2-5 min that leads to the Ga(In)N formation. Next stage is an increasing of temperature up to 1000 °C without ammonia flux. Appearing of GaN QDs was observed in temperature range of 900-930 °C. The RHEED patterns of the main stages of GaN QDs formation are shown in Figure 2.

Kinetics of GaN QDs formation is shown in Figure 3. The kinetic curve shape also demonstrates S-like character as in Figure 1, consequently this process should be treated as phase transition. In Figure 3 the μ denotes the chemical potential of quantum dot, ε – is the bonding energy of the QD with the surface, T – surface temperature and k is the Boltzmann constant. The value of μ increases with time because of increasing of entropy part of the chemical potential as the QDs are accumulated on the surface. The experimental kinetic curve of GaN QDs formation in fact represents a QDs density dependence on chemical potential μ, so this is an isotherm of the phase transition.

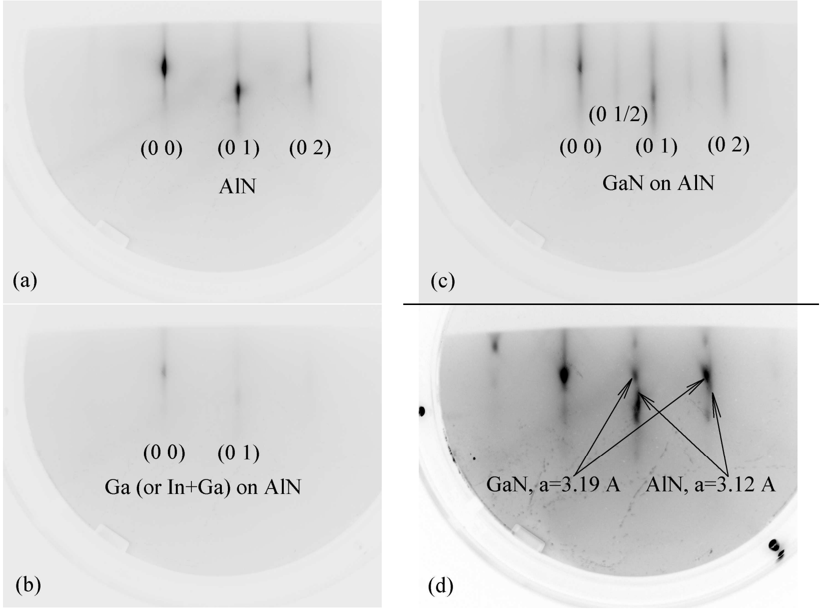

Figure 2. RHEED patterns observed at different stages of GaN QDs formation: (a) pattern of the initial AlN surface, (b) pattern appeared after Ga metal deposition, (c) pattern of the 2D GaN layer under ammonia flux, (d) pattern observed at point of 2D-3D transition during heating of the substrate from 540°C till 1000°C.

The experimental isotherm was described in the frame of lattice gas model for the first-order phase transitions. For calculations we have used the equation (6) on page 137 of the Reference [6]. Obviously, the experimental curve can not exactly follow for the discontinuous transition; nevertheless satisfactory agreement between theoretical and experimental curves is clearly seen.

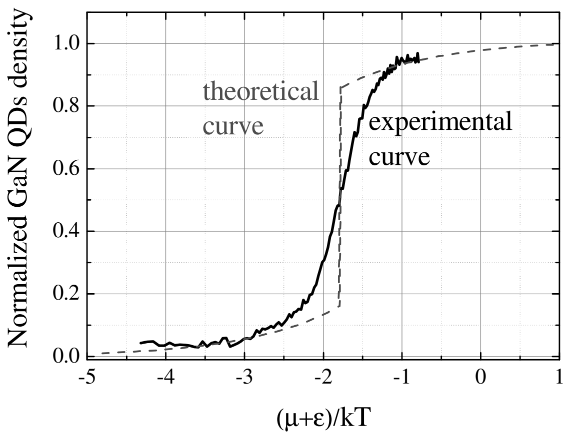

Figure 3. Experimental and theoretical isotherms of GaN QDs formation at T=900 °C.

Density of the GaN QDs was found about 10^8 cm^{-2}, as it confirmed by micro-photoluminescence (micro-PL) spectra obtained from single quantum dot. The sample for micro-PL measurements was fabricated by overgrowth of the prepared GaN QDs ensemble by AlN layer of 100 nm thick. Figure 4 shows micro-PL spectra of the GaN/AlN QDs at different excitation powers. The spectra contain two peaks at 401.2 nm and 403.7 nm with full width at half maximum of about 10 meV. The linewidth is similar to the observed in [7] for the single-QD transitions. The linewidth is larger than lifetime-determined linewidth of the excitonic transition in the single QD. We suppose that the broadening is caused by charge fluctuations on defects near the QDs. The first peak have superlinear power dependence and disappears at low excitation power. Power dependence of the intensity of the PL lines is well described by Poisson-distribution model [8]:

$$I(P) = \frac{I_0}{k!}\left(\frac{P}{P_0}\right)^k \exp\left(-\frac{P}{P_0}\right) \tag{1}$$

With k=1 (exciton) for the line at 403.7 nm and k=2 (biexciton) for the line at 401.2 nm. Thus we attribute the peak at 403.7 nm to the excitonic transition and the peak at 401.2 nm to the biexcitonic transition. Biexcitonic peak is higher in photon energy than the excitonic peak by 19 meV which is in agreement with typical biexcion binding energy in GaN/AlN QDs [9]. The QDs with sufficiently large size have negative biexciton binding energy because of electron and hole wavefunctions are separated by built-in electric field and the interaction between two electrons and two holes is higher than electron-hole interaction [10].

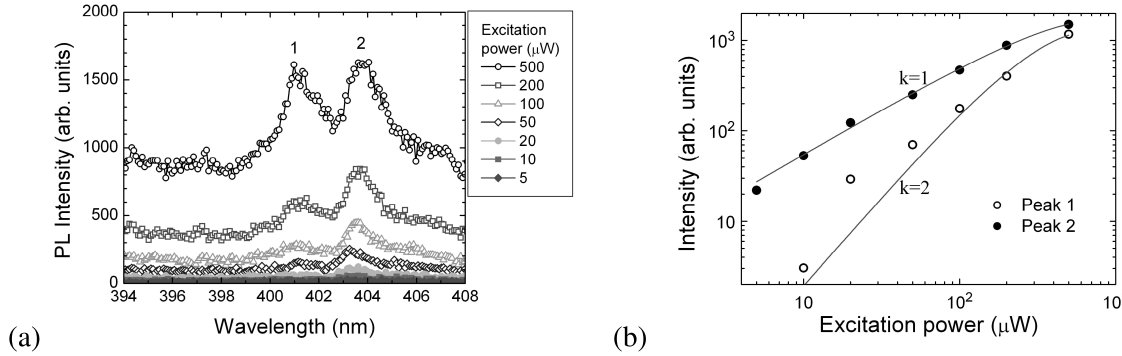

Figure 4. (a) Micro-photoluminescence spectra of GaN/AlN QD structure at different excitation powers. Sample temperature is 4 K. (b) Power dependence of integrated intensities of the PL lines (circles – experiment, solid lines – fitting by equation (1)).

4. Conclusion

Original method of Ga(In)N/AlN quantum dot formation by ammonia MBE have been developed. The method allows obtaining high-quality QDs with density in the range 10^7-10^9 cm^{-2}. PL lines corresponding to excitonic and biexcitonic transitions of single QDs were observed.

Acknowledgments

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