Fabrication of GaN Nanostructure Using Metalorganic Chemical Vapor Deposition

B.Shen, P.Chen, M.Wang, Y.G.Zhou, Z.Z.Chen, L.Zhang, and Y.D.Zheng

Department of Physics and National Solid Microstructure Lab, Nanjing University, Nanjing 210093, P.R.China

E-mail: ydzheng@nju.edu.cn

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Abstract

Nanometer-scale GaN dots on sapphire substrates have been successfully fabricated using metalorganic chemical vapor deposition (MOCVD) with two-step processing, including depositing at temperatures around 500℃ and annealing at 1050℃. The size of GaN dots is around 40 nm in diameter, and the density is from 5×10^8 to 6×10^9 cm^{-2}, depending on the duration and temperature of the deposition as well as of the annealing. GaN dots only form after annealing at high temperatures, which is explained that the initial layer deposited around 500℃ is a high energy intermediate phase in which the large strain energy is not relaxed.

I. Introduction

The fabrication of nanometer-sized materials has gained considerable attention because of their potential uses in both mesoscopic research and the development of nano-devices[1,2]. Low-dimensional structures of quantum dots and wires bring lots of new physical phenomena and new technology on the application of devices. The III-V nitrides with wide energy gap, such as GaN, AlGaN, have been successfully used in blue light emitting, laser diodes and other devices[3,4]. Recently, the fabrication and characterization of GaN quantum dots and wires have been reported by several research groups[5,6]. Similar to the devices fabricated with Si and other III-V semiconductors, reducing the dimension of the active layer may greatly improve the quality and the application of GaN-based devices.

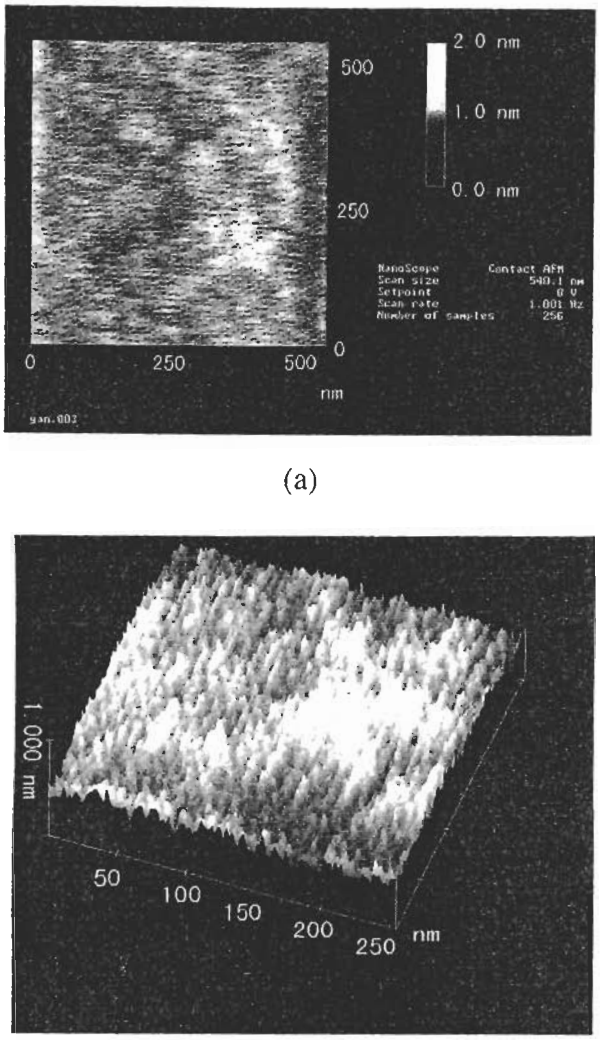
According to Stranski-Krastanow (SK) growth model[7], if the lattice mismatch exists between epilayer and substrate, the initial growth of the epilayer is layer by layer, then the island growth occurs due to the releasing of a large strain in thicker epilayer. The fabrication of quantum dots of Ge, Si and III-V compounds using CVD or MBE techniques is based on this model[8,9]. Usually, in the growth of III-V compounds quantum dots, an initial cladding layer is first grown on substrate, and then quantum dots are grown on the layer surface during a very short time, typically 3 to 10 seconds. Obviously, the short time growth brings some difficulties in controlling the size and the density of dots.

The idea of a system progressing through a succession of phases to reach the final equilibrium phase is well known[10]. If the direct formation of an equilibrium phase has a large kinetic barrier, it may be easier to reach the last state through intermediate phases, each of which has a lower barrier for the formation.

In the present study, we firstly report the successful fabrication of GaN quantum dots using MOCVD with two-step processing, including GaN depositing at temperatures around 500℃ and annealing at 1050℃. By the annealing process, GaN quantum dots are formed in several minuets. This makes the size and the density of the dots to could be controlled more easily in comparison with the previous reports.

II. Experimental

The GaN quantum dots were grown on α-Al_2O_3 (0001) substrate, oriented along [0001] direction. A MOCVD system with a horizontal quartz reactor was used. Before growth, the substrates were cleaned in hot H_2SO_4 and H_3PO_4 (3:1) solution. Then, the substrate was annealed at 1100℃ under H_2 atmosphere for 15 minutes and NH_3 for 5 minutes. As the first growth step, thin GaN layer was deposited at 500℃for 2 to 3 minutes. The growth used a TMG flux of 20 μmol/min, a NH_3 flow of 1 sl/min, and a H_2 flow of 0.6 sl/min. The reactor pressure was maintained at 76 torr. Then, as the second growth step, GaN layer was annealed at 1050℃for 5 minutes under NH_3/H_2 atmosphere to form GaN quantum dots.



(a)

(b)

Fig.1 AFM images of the surface of thin GaN layer deposited on α-Al_2O_3 substrate at 500℃for 2 min. (a) plane view; (b) bird view. The sample is not annealed at high temperatures.

All of specimens were observed using atomic force microscopy (AFM) after the first growth step or the second annealing step. The atomic force microscope is Nanoscope IIIa, Digital Instruments. The density and the size of GaN dots were determined by means of AFM observations.

Ⅲ. Results and Discussion

Fig.1 shows the AFM images of the surface of a GaN thin film deposited at 500℃ for 2 minutes on α-Al_2O_3 substrate. Fig.1a is the plane view, which indicates that GaN thin films prior to annealing at high temperatures has a very plain surface with a roughness less than 0.5 nm. Fig.1b is the bird view, which displays that mini island lines exist, and the orientations of the lines cross an angle of 60 or 120°. Considering the orientation of the substrate, the angle is thought as the result that the islands row up along the orientation of the substrate.

After annealing at 1050℃for 5 minutes, AFM image shown in Fig.2 exhibits that GaN nano-scale dots have been formed. The dots with a diameter of~40 nm and a height of~6 nm are clearly observed and the density is about $5x10^8$ cm^{-2}. This observation indicates that the dots are formed by the annealing at high tempera-tures, which is different from the previous reports on quantum dot growth.

The size and the density of GaN dots could be controlled by changing the duration and temperature of the deposition as well as of the annealing. It has been observed if the sample has the initial GaN film depositing at 550℃for 3minutes and then annealing at 1050℃ for 5 minutes, the density of the dots increase $5x10^9$ cm^{-2} and the height is 10nm. Comparing with the direct deposition of nano-scale dots of GaN and other III-V compounds where a short growing duration less than 10 seconds is required[5,8], both the depositing and annealing are performed with a longer duration in the present work, which is profitable obviously to control the size and the density of GaN dots.

It is well known that there are three models of epitaxial growth: (1) the Frank-Van der Merwe (FVdM) model describing the lay-by-lay growth (2D)[11], (2) the Volmer-Weber (VW) model describing the islands growth (3D) [12], and (3) the SK model describing the lay-by-lay plus the islands growth[7]. The interfacial free energy and the lattice mismatch determine which growth model occurs in a given system. If $\sigma_2 + \gamma_{12} < \sigma_2$, the island growth will occur, where σ_2 is the epilayer surface energy, γ_{12} the interface energy and σ_2 the substrate surface energy.

Due to the large mismatch between GaN epilayer and α-Al_2O_3 substrate, the critical thickness of GaN strain layer is very small. In our experiments, the thickness of the initial GaN layers is about 15 to 20 nm, which is much thicker than the critical one. However, as shown in Fig.1a, a plain surface of the sample after the first deposition step is observed clearly. This indicates that the initial GaN layer is not an equilibrium phase. During the GaN deposition at low temperatures, the low thermal kinetic energy holds back the motion

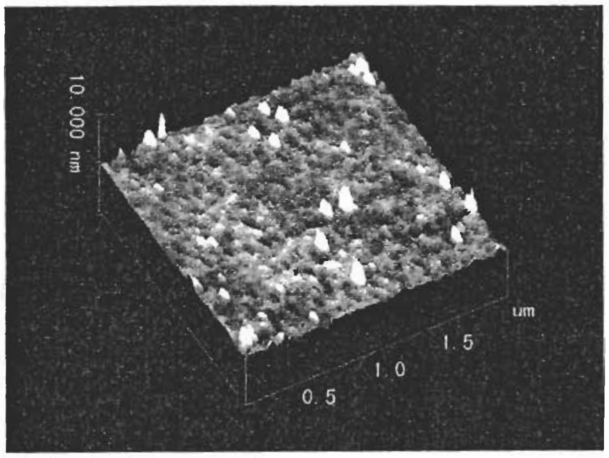


Fig.2 AFM image of GaN quantum dots on α-Al_2O_3 substrates. The sample is performed with GaN depositing at 500℃for 2 minutes and annealed at 1050℃for 5 minutes.

of the atoms or the mini islands. When the sample is annealed at temperatures higher than 1000℃, the mini GaN islands with high thermal kinetic energy would accumulate to some big islands to form GaN dots, while the strain energy is relaxed, and thus the system reaches a final equilibrium phase. In our experiments, additionally, the state of GaN layer performed the annealing at high temperatures but not reaching the final equilibrium phase has also been observed. The AFM image of the GaN layer annealed at 1050℃only for 1.5 minutes is shown in Fig.3. Because the duration of the annealing is not enough, both quantum dots and the mini islands are observed.

Ⅳ. Conclusion

We have successfully fabricated GaN quantum dots on sapphire substrate using MOCVD technique with a two-step processing, including depositing at temperatures around 500℃ and annealing at 1050℃. AFM observations clearly show that the diameter of GaN dots is around 40nmand the density is from 5x10^8 to 6x10^9 cm^{-2}. The size and density of GaN dots are controllable by changing the duration and the temperature of the first deposition step as well as the second annealing step. GaN dots only form after annealing at high temperatures, which is explained that the initial layer deposited around 500℃is a high energy intermediate phase in which the large strain energy is not relaxed.

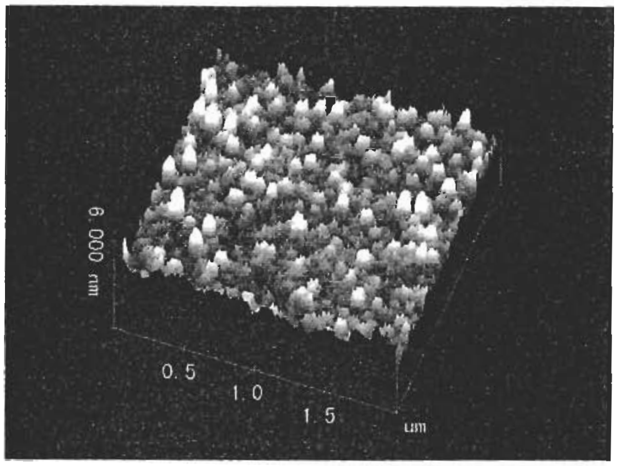


Fig.3 AFM image of GaN initial layer annealed at 1050℃for 1.5 minutes. Both GaN quantum dots and the mini islands are observed.

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