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Self-assembled growth of GaN nanowires

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Abstract. GaN nanowires (NWs) have been grown on Si(111) substrates by plasma-assisted molecular beam epitaxy. The morphology and optical properties of the NWs are influenced by the growth parameters as investigated by the scanning electron microscope. The nucleation process of GaN-NWs is explained in terms of nucleation density and wire evolution with time. The wire length in the nucleation stage shows a linear time dependence. The wire density increases rapidly with time and then it saturates. We explain GaN-NWs growth by making use of the diffusion-induced (D-I) mechanism that explains the dependence of the length on wire diameter for a deposition time longer than the nucleation stage.

1. Introduction

III-nitride semiconductor nanowires (NWs) have been investigated in recent years as potential candidate to fabricate nanoelectronic devices [1, 2]. And they have been grown successfully on different substrates with high crystalline quality and strong luminescence efficiency using plasma-assisted molecular beam epitaxy (MBE) [3, 4]. In this paper, we demonstrate the successful fabrication of GaN NWs on Si(111) under different growth conditions and investigate their optical properties. The growth mechanism is also briefly discussed using diffusion-driven model.

2. Experimental

GaN NWs have been grown on Si(111) under nitrogen-rich conditions by radio-frequency plasma-assisted MBE. A detailed description of various experimental growth conditions and the influence of different parameters viz. substrate temperature, Ga beam equivalent pressure (BEP) and nitrogen (N_2) flow rate can be found elsewhere [3]. The growth morphology and cathodoluminescence (CL) spectroscopy are investigated by a LEO 1550 scanning electron microscope (SEM) equipped with a Zeiss VIS grating monochromator.

3. Results and Discussions

3.1. Nanowire morphology

Growth parameters such as III-V ratio and substrate temperature greatly influence the surface morphology of these NWs. To investigate this, a series of samples are grown on Si(111) substrates by changing only one parameter at a time. In Fig. 1, SEM micrographs of samples grown for 2 hrs. at constant Ga BEP and N_2 flow rate (Ga BEP of 3×10^{-8} mbar and 4 sccm N_2 with 500W power) but at different substrate temperature are shown. A rough surface morphology is observed at 700 °C and some wires start to grow (Fig. 1(a)). A further increase of temperature

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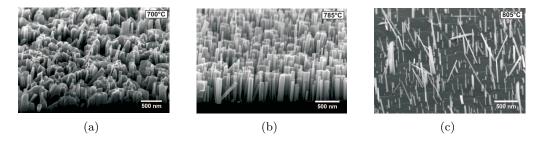


Figure 1. Cross-sectional SEM images of GaN NWs at different substrate temperature as shown in the inset. Ga BEP is 3×10^{-8} mbar and N₂ flux is 4 sccm with 500W power.

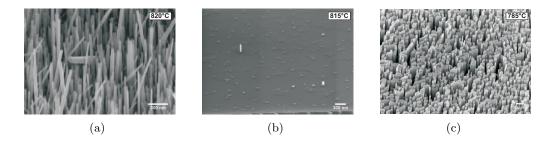


Figure 2. SEM micrographs of GaN NWs at different Ga BEP and N_2 flow rate. (a) Ga BEP: 1.5×10^{-7} mbar and (b) Ga BEP: 3×10^{-8} mbar with 4 sccm N_2 . (c) N_2 flow rate: 2.7 sccm with 500W power and Ga BEP: 3×10^{-8} mbar. A bit more coalescence is visible in NWs as compare to Fig. 1(b) which has been grown at N_2 flow rate of 4 sccm. The growth temperatures are shown in the inset

to 785 °C shows an homogeneous distribution of NWs on the surface (Fig. 1(b)) and finally the density is reduced at 805 °C (Fig. 1(c)). Further increase of temperature leads to growth suppression. As Ga desorption becomes more significant at higher temperature, NW density decreases or almost no growth takes place and this can be compensated by increasing the Ga flux. SEM images of NWs grown at higher Ga flux $(1.5\times10^{-7} \text{ mbar})$ as compared to Fig. 1 are shown in Fig. 2 . Even at 820 °C, many wires are grown (Fig. 2(a)), whereas no growth is observed almost at the same temperature with lower Ga flux (Fig. 2(b)). On the other hand, nitrogen flux also influences the growth as can be seen in Fig. 2(c). Samples show more coalescence at lower nitrogen flux than the higher one. So it is important to keep the effective III-nitride ratio smaller than one to initiate NW growth.

3.2. Growth mechanism

The growth mechanism of GaN NWs by MBE is still not deeply investigated as far as catalyst-free approach is concerned. Growth parameters influence strongly the surface morphology of the nanowires as shown in the previous section and it is difficult to explain the growth mechanism using a general model.

To illustrate this phenomenon using a simplified model, nanowire length and diameter as a function of time can be taken into account in order to determine vertical and lateral growth of these NWs. A series of samples have been grown at different growth time (Figures not shown here) and a detailed description can be found in [5]. The nanowire density goes on increasing up to 60 min of deposition time and then decreases due to the coalescence effect (Fig. 3(a)). The vertical and lateral growth shows linear time-dependent behaviour considering the average of the NWs with similar diameter (Figs. 3(b)). The linear fit for diameter shows probable nucleation

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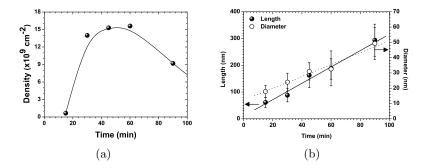


Figure 3. (a) GaN NW density as a function of time. (b) Length and diameter distribution as a function of deposition time shows linear characteristics.

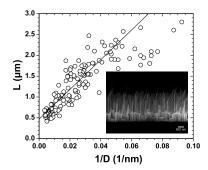


Figure 4. Length and Diameter dependence of GaN NWs along with the fitted function using Eq. 1. The inset shows a typical SEM image of analysed NWs grown for 6 hrs.

diameter of ~ 15 nm before the growth takes place. It must be noted here that this post-growth analysis does not reflect the nucleation time for every single nanowire. Also, the resolution limit of the SEM should be taken into account.

An interesting relation between length (L) and diameter (D) for growth time longer than 360 min can be evaluated. There is an increase of nanowire length for smaller diameter and vice versa as illustrated in Fig. 4. The above mentioned length-diameter dependence can be expressed as

$$L = C_1 \left(1 + \frac{C_2}{D} \right) \tag{1}$$

where C_1 and C_2 are the constants related to the analysed sample [6]. A fit to the experimental data with Eqn. (1) provides the values of 470 nm and 80 nm for C_1 and C_2 respectively. The value of C_1 is related to the thickness of a compact layer and C_2 is related to the diffusion-induced (D-I) growth. If the diffusion length of adatoms is less than the column length then a strong tapering effect is expected. But no substantial tapering effect is observed for NWs with 2-3 μ m length. So this growth mechanism can not related to a shorter diffusion length. Further, it can be concluded that D-I growth below 80 nm (C_2) dominates for thinner columns, whereas adsorption at the tip of the NW (i.e. direct impingement) is the major contributing mechanism for the growth for larger diameter (>80 nm). A very high aspect ratio of about 250 has been achieved for thin, long NWs.

3.3. Optical properties

In order to investigate the optical properties of NWs grown at different substrate temperature, cathodoluminescence (CL) spectroscopy is performed at 10 K. The samples have been grown at

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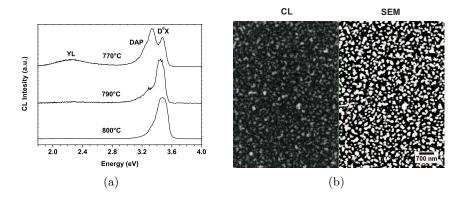


Figure 5. (a) CL spectra of GaN NWs at different growth temperature. Ga BEP is 2.7×10^{-8} mbar and N₂ flor rate is 4 sccm with 500W power. (b) Panchromatic CL and SEM micrographs of the sample grown at 790 °C.

fixed Ga and N₂ flux. CL spectra of these samples are shown in Fig. 5(a). Three main peaks can be identified: one sharp peak beyond 3.45 eV can be attributed to the donor-bound-exciton (DX) transition, the second peak at 3.28 eV is identified as donor-acceptor-pair (DAP) and the broad peak centred at 2.2 eV is known as yellow-luminescence (YL). Energy variation of DX transition can be due to different amount of strain in NWs grown on Si(111). A strong decrease of YL band as well as DAP emission with increasing substrate temperature indicates that NWs grown at higher temperature have higher crystalline quality than the ones grown at lower temperature. Top-view Panchromatic CL image of NWs grown at 790 °C shows the luminescence centred at the column position (Fig. 5(b)).

4. Conclusions

GaN NWs have been fabricated successfully on Si(111) by MBE. The effect of different growth parameters on the morphology of the NWs has been investigated. A probable nucleation diameter of ~ 15 nm has been estimated from the diameter-time dependent relationship. An inverse dependence of the NW length on its diameter exists which has been explained by D-I growth mechanism. CL spectra show higher crystalline quality of NWs grown at higher deposition temperature.

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