Supporting Information

Heteroepitaxial Growth of Vertically-Aligned GaN Single-Crystalline Microrod Arrays on Silicon Substrates

Authors
   Chun-Wei Chuang† and Franklin Chau-Nan Hong*,†

Affiliations
   †Department of Chemical Engineering, National Cheng Kung University, 1 University Road, Tainan 70101, Taiwan

*Corresponding author
   Franklin Chau-Nan Hong
   E-mail: hong.franklin@gmail.com
1. Crystalline Quality and Normalization of Photoluminescence Spectra

In this study, the crystalline qualities of gallium nitride (GaN) microrod arrays were evaluated via the measurement of the room-temperature photoluminescence (PL) with using the helium-cadmium laser (He-Cd laser) of 325 nm as the excitation light source. In the meantime, the material properties of GaN were characterized in the PL spectra. For instance, the peak of the near-band-edge emission (NBE emission) of GaN is at the position about 363 nm and the yellow luminescence (YL), caused by crystal defects, can be shown in the range of 510 nm to 700 nm. During the PL measurement, the carriers inside the GaN crystals can be excited by the incident light energy from the 325-nm laser and further react with defects and grain boundaries, which will produce not only the YL but also the non-radiative recombination. In addition, the luminescence efficiency of GaN will be reduced due to the consumption of carriers inside materials at the positions of defects and grain boundaries. Therefore, the formation of point defects, dislocations and grain boundaries should be avoided for the growth of vertically-aligned GaN microrod arrays in order to obtain the high luminescence efficiency.

Apart from judging the YL of material defects in the PL spectra, the determination of crystalline qualities should also consider the little bias for the light intensity of the incident light source because the intensities of NBE emission and YL both increase (or decrease) with the enhanced (or weakened) laser light for the PL measurement in different time periods. For a material with a lower intensity of YL, its intensity of YL will be increased to a higher value when the incident laser light becomes stronger, causing some errors in the comparison of crystalline qualities. Besides, the situation of GaN NBE emission is similar to that of YL when evaluating the crystalline quality of GaN. Therefore, YL and NBE emission should both be taken into account in the more accurate comparison of crystalline qualities, and their intensity values in the PL spectra should be utilized simultaneously to represent the crystalline qualities of materials.

The lower intensities of YL or the higher intensities of NBE emission stand for the better crystalline qualities of GaN microrod arrays. Thus, the intensity ratio of YL to NBE emission (YL/NBE emission) is employed to decide the level of crystalline qualities for different samples of vertically-aligned GaN microrod arrays. So as to compare the intensity ratio of YL to NBE emission more easily, the normalization treatment was applied to the PL curves. The PL spectra for the GaN grown at the three different temperatures, as shown in Figure S1(a), are used as the demonstration to explain the normalization treatment. In the PL spectra, the normalization is to define the intensity value of NBE emission of GaN as one for each PL curve so that all the y-axis values change to the values in the range of zero to one as shown in Figure S1(b). In the meanwhile, the differences in the crystalline qualities can further be found via the direct observation on their YL intensities of the normalized PL curves, owing to the same value of NBE emission after the normalization treatment.
Figure S1. PL spectra for the vertically-aligned GaN microrod arrays grown at the temperatures of 940 °C (black curve), 950 °C (red curve) and 960 °C (green curve). (a) The original linear curves before the normalization. (b) The linear curves after the normalization. (c) The normalized curves on the logarithmic scale.

After taking the logarithm of the three normalized PL curves, the intensity values of the curves do not go to zero in the long-wavelength region as shown in Figure S1(c). Moreover, the background values as the noise signals in the PL spectra are relatively low because the crystalline qualities of the vertically-aligned GaN microrod arrays are not poor. The thermal noise as the background signal is mainly related to the crystalline qualities of the measured materials and the equipment of the PL measurement. Before the correct PL measurement in the darkroom, cooling down the temperature to lower than 140 K by adding the liquid nitrogen is important in order to avoid the interference of the thermal noise, derived from the environment, during the measurement. Therefore, the reason why the obvious thermal noise appears on PL curves of GaN microrod arrays is that the materials have the much poor crystalline qualities instead of the factors of the equipment and environment under the normal PL measurement.
2. Steps for Analyses of Materials and Confirmation of Characteristics

After completing the experiments of crystal growth, the following steps from (1) to (4) were carried out to analyze the morphology, the material type, the crystallization manner and the crystalline quality step by step.

(1) Morphology: vertically-aligned microrod arrays

The morphology, the sizes and the arrangement manner for the as-grown materials were observed by using the scanning electron microscopy (SEM), which verifies the morphology of vertically-aligned microrod arrays with sub-micron sizes.

(2) Material type: GaN

For the microrod arrays, the items of the material type, the crystallinity and the preferred crystal plane were analyzed via the X-ray diffraction (XRD). The result shows that the material is high-crystalline GaN with the (002) crystal plane.

(3) Crystallization manner: single-crystalline GaN

Furthermore, the crystals were affirmed to be the single-crystalline GaN with the [0001] growth direction via the transmission electron microscopy (TEM) analysis and the corresponding selective area electron diffraction (SAED). By summarizing the results of (1) to (3), we can confirm that the analyzed material in this study is the vertically-aligned GaN single-crystalline microrod arrays.

(4) Crystalline quality: high crystalline quality

After the steps of (1) to (3), the room-temperature PL was employed to determine the crystalline qualities of the vertically-aligned GaN single-crystalline microrod arrays. The improvement in the crystalline quality and the appearance of the high crystalline quality can be revealed in the PL spectra, and their related details are reported in the article.