Growth of InGaN self-assembled quantum dots and their application to photodiodes

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Nanometer-scale InGaN self-assembled quantum dots (QDs) have been prepared by growth interruption during metalorganic chemical vapor deposition growth. With a 12 s growth interruption, we successfully formed InGaN QDs with a typical lateral size of 25 nm and an average height of 4.1 nm. The QD density was about \(2 \times 10^{10} \text{cm}^{-2}\). In contrast, much larger InGaN QDs were obtained without growth interruption. InGaN metal-semiconductor-metal photodiodes with and without QDs were also fabricated. It was found that the QD photodiode with lower dark current could operate in the normal incidence mode, and exhibit a stronger photoresponse.

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I. INTRODUCTION

III–V nitride semiconductor materials have a wurtzite crystal structure and a direct energy band gap. At room temperature, the band gap energy of AlInGaN varies from 0.7 to 6.2 eV depending on its composition. Therefore, III–V nitride semiconductors are particularly useful for light-emitting devices and photodetectors in this wavelength region. Indeed, III–V nitride-based blue and green high brightness light-emitting diodes (LEDs)\(^1\),\(^2\) made from InGaN/GaN quantum well (QW) structures are now commercially available for traffic light source and full color display. However, relatively few nitride-based blue/ultraviolet (UV) photodiodes could be found in the literature as compared to nitride-based LEDs. Blue/ultraviolet (UV) photodiodes are important devices that can be used in various commercial and military applications. For example, these devices can be applied in space, medical, and environmental fields. Currently, light detection in the blue/UV region still uses Si photodiodes. However, since room temperature band gap energy of Si is only 1.2 eV, the responsivity of Si photodiodes is low in the blue/UV region. With the advent of optoelectronic devices fabricated on wide direct band gap materials, it becomes possible to produce high performance solid-state photodiode arrays sensitive in the blue/UV region. Depending on device structure, nitride-based p–n junction diode,\(^3\) p–i–n diode,\(^4,5\) p–p–n diode,\(^6\) Schottky barrier detector,\(^7\) and metal-semiconductor-metal (MSM) photodiodes\(^8\)–\(^10\) could all be used to detect blue/UV signal. Among these devices, MSM photodiodes have an ultralow intrinsic capacitance and their fabrication process is also compatible with field-effect-transistor (FET)-based electronics. Thus, one can easily integrate GaN MSM photodetectors with GaN FET-based electronics to realize a nitride-based optoelectronic integrated circuit.

Low-dimensional carrier confinement nanostructures such as quantum wires and dots (or islands) are quite attractive for application to high-performance electronic and optical devices. Recently, it has been shown that nitride quantum dots (QDs) can be self-organized using the strain-induced Stranski–Kranstanov growth mode.\(^11\)–\(^13\) It has also been shown that nitride QDs can be self-organized using growth interruption during metalorganic chemical vapor deposition (MOCVD) growth.\(^14\) Although the successful fabrication of InGaN QDs has been reported by many research groups, very few reports of InGaN QD-based optoelectronic devices could be found in the literature.\(^15\)–\(^16\) In this article, we use MOCVD growth interruption to form nitride self-assembled quantum dots (SAQDs), InGaN QD MSM photodiodes were subsequently fabricated. The photoluminescence (PL) properties of InGaN QDs and the current–voltage (I–V) characteristics of fabricated MSM photodiodes will be discussed.

II. EXPERIMENTS

Samples used in this study were grown on (0001)-oriented sapphire (Al\(_2\)O\(_3\)) substrates in a vertical low-pressure MOCVD reactor with a high-speed rotation disk.\(^17\),\(^18\) The gallium, indium and nitrogen sources were trimethylgallium, trimethylindium, and ammonia, respectively. After a 30-nm-thick low-temperature GaN nucleation layer was deposited onto the sapphire substrate at 500 °C, the temperature was raised to 1000 °C to grow a 2-μm-thick undoped GaN buffer layer. The growth temperature was then reduced to 730 °C to grow InGaN QDs with growth rate of 0.04 nm/s. During the...
deposition of InGaN, an interrupted growth method was employed in preparing sample A. In other words, we deposited a 1.2-nm-thick InGaN layer on top of the undoped GaN buffer layer, stopped the growth for 12 s, and then deposited another 1.2-nm-thick InGaN layer so as to achieve a total InGaN layer thickness of 2.4 nm, as shown in Fig. 1. For comparison, sample B was prepared by directly depositing a 2.4-nm-thick InGaN layer on top of the GaN buffer layer, as also shown in Fig. 1. Furthermore, samples C and D without the QDs were also prepared by depositing a 2.4-nm-thick and 100-nm-thick InGaN layer on top of the undoped GaN buffer layer, respectively. It should be noted that these four kinds of samples have the same average indium composition. Room temperature surface morphologies of the InGaN nanostructure samples were then characterized ex situ by an atomic force microscopy (AFM) system (Shimmadzu SPM-9500IZ) with a sharpened Si$_3$N$_4$ tip. PL was also used to study the optical properties of these samples under room temperature. During PL measurements, a 325 nm He-Cd laser was used as the excitation source. The collected luminescence signal was dispersed by a monochromator, and detected by a photomultiplier tube (PMT).

InGaN QD MSM photodiode was then fabricated on sample A as photodiode I. For comparison, 2 MSM photodiodes without QDs were fabricated on samples C and D as photodiodes II and III, respectively. Figure 2 shows the schematic structure of the InGaN QD MSM photodiodes used in this study. The fabrication process of these InGaN MSM photodiodes was as follows. Prior to the deposition of contact electrodes, wafers were dipped in a diluted hydrochloric acid water solution (HCl:H$_2$O=1:1) for 3 min to remove native oxides. A Ni layer was subsequently deposited onto the sample surface by e-gun evaporator to serve as the metal contact. Standard lithography and etching were then performed to define the interdigitated contact pattern. The fingers of the contact electrodes were 10 µm wide and 200 µm long with a spacing of 10 µm. The active area of the whole device was 200×200 µm$^2$. Finally, Au layers were then deposited on top of the contact electrodes to serve as bonding pads. A HP-4155B semiconductor parameter analyzer was then used to measure the current–voltage ($I$–$V$) characteristics of these MSM photodiodes in dark and under illumination. For photocurrent measurements, a 150 mW gas Ar laser with 457, 465, 476, 488 and 514 nm line, illuminating from the front side of the fabricated photodiodes, was used as the light source.

III. RESULTS AND DISCUSSIONS

Figures 3(a) and 3(b) show 500–500 nm$^2$ two-dimensional (2D) AFM images of samples A and B, respectively. As shown in Fig. 3(a), it can be seen that small circular InGaN SAQDs were formed by the interrupted growth mode. From these AFM pictures, it was found that the diameter of these circular QDs was in the range of 20–38 nm, with an average height of 4.1 nm. On the other hand, the density of these QDs was estimated to be around 2 $10^{10}$ cm$^{-2}$. In contrast to the small circular QDs observed from sample A, large oval InGaN islands were found in sample B without growth interruption. It was found that the longer width of these oval islands was around 140 nm while the shorter width was about 70 nm, with an average height of 1.7 nm. We also found that the density of these large oval islands was about $3.5\times10^8$ cm$^{-2}$. From these AFM pictures shown in Figs. 3(a) and 3(b), it can be seen clearly that by introducing the interrupted growth method, we could significantly change the surface morphology of the MOCVD grown InGaN samples. It should be noted that the size of the nanostructures observed from sample A was much smaller than that observed from sample B. As a result, growth interruption could release the partial strain energy of InGaN epitaxial layer when InGaN was grown more than the critical thickness 0.8 nm, i.e., 1.2 nm >0.8 nm, then continued growing another 1.2 nm of InGaN, we can realize the nanoscale and discrete SAQDs. Thus, we should be able to observe a more significant quantum confinement effect from sample A.

Figure 4 shows the measured PL spectra for samples A, B and D. It can be seen that the PL peak position of sample 1 is located at 2.645 eV while the PL peak position of sample 2 is located at 2.578 eV. It was also found that the normalized PL intensity observed from sample 1 was 50% larger than that observed from sample 2. In other words, the introduction of growth interruption would result in a PL blueshift as large as 67 meV. It should be noted that the nominal thickness of InGaN epilayer of these two samples was the same (i.e., 2.4 nm), i.e., they had the same InGaN coverage. Hence we should attributed the huge PL blueshift to the lateral size
effect of nanostructures. It is well known that the subband transition energies will increase when the size of nanostructure becomes smaller. Thus, although the InGaN coverage was the same for samples A and B, we could observe a larger PL transition energy from sample A prepared with growth interrupt, since growth interrupt will result in a smaller nanostructure size. Furthermore, we can see that the sample A is about 180 times the normalized PL intensity of sample D. It is possible that sample D has no quantum structures like quantum wells or dots to confine the exitons which will be trapped by defects (e.g., threading dislocations) which results in the low probability of radiation recombination. Hence sample D showed weak PL intensity.

Figures 5(a) and 5(b) show photocurrent and dark current of InGaN MSM photodiodes I and II fabricated on samples A and C, respectively. It can be seen that photocurrent and dark current both increase slowly as the applied bias increases. With the same device size, it was found that photodiode I had slightly lower measured dark currents than photodiode II. On the other hand, it was found that the photocurrent observed from photodiode I was much larger than that observed from photodiode II. It should be noted that these photodiodes were illuminated normally. Due to the special structure for samples A and C having nanoscale QDs and thin film, respectively, we can treat the photodiodes I and II as a QD photodetector and a QW photodetector. As a result, for the sake of the polarization selection rules, QW (2.4-nm-thick InGaN) detectors which are not sensitive to radiation that is incident perpendicular to the QW, on the contrary, the QD detector can operate in the normal incidence mode showing better photoresponse.

Figure 6 shows photocurrent to dark current contrast ratio of these three photodiodes. With a 10.1 V applied bias, it was found that photocurrent to dark current contrast ratio equals 400, 11, and 31, respectively. The large photocurrent to dark current contrast ratio observed from photodiode I could
again be attributed to the formation of nanoscale InGaN SAQDs by growth interruption. It is possible that InGaN QD as a good quantum capture system will get more photogenerated carriers under such illumination (blue-green light), hence the photocurrent easily increases when an external bias (electric field) was applied onto the device. On the other hand, the 100-nm-thick InGaN could merely provide less effective photogenerated carriers under illumination, exhibiting poor photoelectric performance. As a result, we could observe a larger photoresponse from photodiodes with InGaN nanostructures.

IV. CONCLUSION

In summary, it has been demonstrated that we can use interrupted growth method in MOCVD to fabricate InGaN SAQDs. With a 12 s growth interruption, we successfully formed InGaN QDs with a typical lateral size of 25 nm and an average height of 4.1 nm. The QD density was about 2 \times 10^{10} \text{ cm}^{-2}. In contrast, much larger InGaN nanostructures were obtained without growth interruption. InGaN MSM photodiodes with and without QDs were also fabricated. It was found that the InGaN QD photodiode with lower dark current can operate in the normal incidence mode. We could achieve a much larger photocurrent to dark current contrast ratio from MSM photodiodes with nanoscale InGaN SAQDs.

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