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Development of substrate-removal-free vertical ultraviolet light-emitting diode (RefV-LED)

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A vertical ultraviolet (UV) light-emitting diode (LED) that does not require substrate removal is developed. Spontaneous via holes are formed in n-AlN layer epitaxially grown on a high conductive n++Si substrate and the injected current flows directly from the p-electrode to high doped n++Si substrate through p-AlGaN, multi-quantum wells, n-AlGaN and spontaneous via holes in n-AlN. The spontaneous via holes were formed by controlling feeding-sequence of metal-organic gas sources and NH3 and growth temperature in MOCVD. The via holes make insulating n-AlN to be conductive. We measured the current-voltage, current-light intensity and emission characteristics of this device. It exhibited a built-in voltage of 3.8 V and emission was stated at 350 nm from quantum wells with successive emission centered at 400 nm. This UV LED can be produced, including formation of n and p electrodes, without any resist process. © 2014 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [http://dx.doi.org/10.1063/1.4867090]

Ultraviolet (UV) light in the wavelength region from 210 to 400 nm is used for a wide range of medical applications such as sterilization of viruses and bacteria phages, photochemical decomposition, and photosynthesis. To date, mercury lamps and excimer lamps are the main light sources of UV light. However, because of the Minamata Convention on Mercury in the United Nations Environment Program1 the use of mercury lamps is being phased out. AlGaN base UV light-emitting diodes (LEDs) have been recently developed as an alternative to mercury and excimer lamps in the UV light region.2–4 UV LEDs can emit light at any wavelength desired in the DUV and UV region. However, AlGaN base DUV and UV LEDs are still used infrequently because of their high price and limited power. One of the reasons it is difficult to produce high-power UV LEDs is the heating of the device because of inefficient light emission. To encourage the use of UV LEDs, it is very important to realize high power operation with a low production cost. Herein, we report a new vertical UV LED which will make possible high power operation in future, the simple device fabrication process and new crystal growth technology for the device. A vertical-type UV LED has previously been developed that shows efficient cooling and high-power operation.5,6 However, to realize this vertical UV LED, removal of the substrate is required. Defects are generated on the removed surface of the epitaxially grown crystal during the laser removal process that is commonly used. Especially in the case of UV LEDs, a sacrificial layer should be introduced to remove the epitaxial layer from the substrate.7

Unfortunately, this insertion of a sacrificial layer complicates the epitaxial growth conditions. One alternative to the sacrificial layer is to develop a new technology to realize vertical UV LEDs that do not require removal of the substrate (hereafter referred to as removal-free vertical UV LED,
RefV-UV LED. We have developed a new technology called the spontaneous via-hole formation method. In this method, high-resistivity n-AlN becomes conductive and vertical current flow from p-electrode to n⁺-Si substrate becomes possible. This method shortens the time for crystal growth and simplifies the device fabrication process because removal of the substrate is not necessary and damage resulting from removal is avoided. In addition, n⁺-Si and n-AlN are suitable for device cooling because of their high heat conductivity, resulting in the potential for large-area devices with high power operation.

The device fabrication processes and typical device structures of the RefV-UV LED are shown in Figures 1. An advantage of vertical LEDs compared with horizontal ones is a shorter current pass length of 1–2 μm versus 50 μm of horizontal LEDs. In addition, vertical LEDs are thin, the active layer is close to the heat sink, and cooling efficiency is very high. However, the electrical conductivity of the AlN layer is very low and, after removal of the substrate in conventional vertical LED, the remaining AlN layer should be removed by RF ion etching to leave an n-AlGaN contact layer. However, RF ion etching damages the AlGaN multi-quantum-well (MQW) layer.

In the RefV-UV LED, the epitaxial layer is grown on an n⁺-Si(111) substrate using an alternative doping method and spontaneous via-holes methods.

In the RefV-UV LED using an n⁺-Si substrate, the formation of cracks in the Si-doped n-AlN layer and the n-AlGaN layer, which usually appear because of the high strain in Si-doped AlN and AlGaN, can be avoided using the spontaneous via-hole formation method. As a result, we can produce crack-free Si-doped epitaxial layers of n-AlN and n-AlGaN. To achieve sufficient current flow from the p-electrode to the n⁺-Si substrate in the RefV-UV LED, highly conductive n-AlN is needed. However, using the conventional method, it is difficult to obtain a high carrier concentration of electrons in n-AlN and n-AlGaN. To induce flow of the current in the vertical direction, the n-AlN and n-AlGaN layers need to be thin and the conductivity of the n-AlN and n-AlGaN layers is increased using alternative doping crystal growth and spontaneous via-hole formation methods. In the alternative doping crystal growth, the activation energy of dopant decreases and high carrier concentration can be expected. Spontaneous via holes also enhance the current flow due to the dangling bond inside via holes. Current can flow through this dangling bond. In this case, the n⁺-Si substrate can be used as an n-side electrode. Therefore, etching from the p-side to make an n-contact, which is performed in horizontal LED fabrication, is not necessary and the AlGaN MQWs are not damaged.

In the spontaneous via-hole formation method shown in Figure 2, we first grow an n-AlN buffer layer after Al injection. Another n-AlN layer is then grown using an alternative growth method with a controlled III-V ratio and temperature. During the growth of this layer, spontaneous via holes are formed (Fig. 2). The number of via holes can be controlled by changing the injection time of Al, growth temperature, and the III-V ratio. This means that at the moment of Al injection, Al dots spontaneously form on the n⁺-Si substrate. During the alternative growth of n-AlN at a suitable temperature and III-V ratio, via holes are generated on Al dots up to the end of the growth of n-AlN. That is, spontaneous via holes are formed in an n-AlN buffer layer on the n⁺-Si substrate. The left side of Fig. 3 depicts spontaneous via holes on the top surface of n-AlN. In our experiments, the number of via holes is about 40 per 10 μm². The structure of the dots varies and their size is between 1 and 2 μm. The dots are uniformly distributed over the whole 2-inch wafer substrate.

After the growth of n-AlN, an n-AlGaN layer is grown on it using an alternative feeding method. In alternative n-AlGaN crystal growth, the growth in the horizontal direction is enhanced in some growth condition compared with that of n-AlN and a mirror-like flat surface is obtained (right side of Fig. 3) that does not contain any cracks, even though it is Si-doped AlN and AlGaN.

We carried our direct measurement of the conductance of AlN layer including via holes. The conductance measured was AlN sandwiched between n⁺Si and high doped thin n-AlGaN. Ohmic contacts of both sides are formed. In the experiment the number and size of via holes are controlled by adjusting the Al injection time at the beginning stage of the crystal growth. The area of via holes is changed from 4 × 10⁶ to 16 × 10⁶ μm² / cm². The conductance increases about 5 times by increasing the area of via holes from 4 × 10⁶ to 16 × 10⁶ μm² / cm². The mechanism of the increase of the conductance of AlN is not sure at this moment but from the experimental result of the proportional increase of the conductance by increasing the area of via holes, AlGaN grown on AlN with layer by layer growth
FIG. 1. Fabrication and structure of the RefV-UV LED.
mode (which enhances lateral movement of adsorbed source molecules on the surface) may fill out inside via holes and the via hole may be conductive. The number of carriers is about $10^{16}$/cm$^3$ and the conductance is quite stable before and after the experiment. Details of the characteristics of via holes including crystal growth condition will be reported elsewhere.

The half-width of X-ray diffraction peak for AlGaN is about 800 s for the (002) surface and about 1000 s for the (102) surface. At this point, the half-width for the (102) surface is not sufficiently narrow, but X-ray diffraction measurement observes diffracted X-ray from whole layer of AlGaN including via hole termination layer. In this experiment the quality of AlGaN near the interface of AlN is not good but it becomes better and better approaching the surface. So, it seems the crystal quality at the top surface may be much better. However, it should be improved by controlling the growth conditions.

Usually, electrode formation takes a long time and damages the device surface. For example, if the resist processes are repeated, the crystal is damaged. However, in the RefV-UV LED, the electrode formation process is simple and the resist process is not required. In this LED, the substrate is $n^+$-Si and the electrode is formed by deposition of metal on the back side of the $n^+$-Si substrate just like that in a solar cell. In the case of the p-electrode, metal deposition of Ni and Au through a metal deposition mask without resist can be carried out. The p-GaN layer used as a p contact layer (Fig. 1) is etched using the electrode as a mask, which is a self-aligning method and means the resist process is not necessary. Using this method, the process time is about 20% that of conventional horizontal LED devices.

Current-light intensity ($I-L$) and Current-voltage ($I-V$) characteristics of the RefV-UV LED are shown in Figures 4 and 5, respectively.

$I-L$ characteristics of the RefV-UV LED (Fig. 4) operated under RT CW mode show rapid increase of light intensity against the current injected. The power has not yet been observed but it seems about a few hundred $\mu$W depending on the electrode size.
FIG. 4. I-L characteristics of the RefV-UV LED. The upper inset shows near field emission pattern of this device. The size of this device is 400 μm × 400 μm. The lower inset shows the EL spectrum.

FIG. 5. I-V characteristics of the RefV-UV LED.
The emission spectrum of this device is illustrated in lower inset of Fig. 4. The emission is started at the wavelength of about 350 nm with successive emission around 400 nm. This built up wavelength of the emission corresponds to the emission from the quantum well designed and successive emission in longer wavelength comes from emission of GaN used as p-contact layer excited UV emission from MQW. The oscillation behavior comes from interference of the emitted light between the surface and hetero interface.

The near field pattern of the RefV-UVLED is shown in upper inset of Fig. 4. The size of tip is about 400 micron.

I-V characteristics in Fig. 5 show that the built-in voltage of this device is 3.8 V with a sharp build-up of current against voltage at this point. This built-in voltage is almost same as the band gap of the AlGaN hetero structure used. The backward current break down voltage was 35V. These results suggest good p-n junction is formed. The series resistance of this device was 13.5Ω. This resistance is not enough low at this moment but it will be improved by controlling the crystal growth condition.

In conclusion we have developed RefV-UV LEDs in which the current flows transversely from the p-electrode to the n⁺-Si substrate. For realizing these devices spontaneous via holes method is useful. The device emits the light 350 nm to 450 nm which comes from the emission of quantum wells and related defect centers. The built in voltage was 3.8 V and reverse break down voltage was of 35 V. This built in voltage is almost same as the band gap of the AlGaN used for our devise. This RefV-UV LEDs provides an answer for the problem of substrate removal that is intrinsic to conventional vertical LEDs. The process used to fabricate the RefV-UV LEDs is quite simple and will allow production of large-area and high-power UV LEDs. This simple concept will contribute to expanding the applications of UV LEDs.

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