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ABSTRACT

For the first time, light-emitting diodes based on gallium nitride nanopillar crystals were prepared directly on a multi-crystalline silicon substrate, which is widely utilized in low-cost solar cells. Several double-hetero-p–n-junction structures were fabricated, and bright light emission was obtained from the diodes. In addition, white-light emission was observed in another diode. The multi-crystalline Si substrate can be added to a candidate substrate to realize practical, novel, large-area light-emitting devices.

I. INTRODUCTION

Gallium nitride (GaN) and other group-III nitride semiconductors possess good properties for fabricating high-performance light-emitting devices.[1–4] However, most nitride semiconductor crystals are grown on single-crystal substrates, such as sapphire, to achieve high crystallinity and hence high performance.[5–11] However, if high-quality group-III nitride crystals can be directly grown on various large non-single-crystalline substrates at a low cost, their application to novel large-area light-emitting devices, such as large-area micro-light-emitting diode (LED) displays[12] and/or planar-type illumination devices, will increase considerably.

Recently, the number of reports on the growth of group-III nitride-based LEDs directly on non-single-crystalline substrates has increased, for example, quartz glass[13–16] or several rare-metal substrates,[17–20] although the number is significantly less than that of epitaxially grown single-crystalline LEDs. We have attempted to directly grow GaN-based crystals on several non-single-crystalline substrates.[21–23] As a representative substrate among them, the multi-crystalline Si wafer, which has been widely utilized in low-cost solar cells, presents several advantages for practical applications. It is typically manufactured using the casting method, which allows easy control of its size, surface roughness, and conductivity.

However, the direct fabrication of LEDs on multi-crystalline Si wafers has not been reported. In addition, nanostructured crystals, such as nanocolumns and nanowires, have proven to be suitable for the fabrication of high-performance nitride-based light-emitting devices.[24,25] In this study, GaN nanopillar-based p–n-junction diodes were grown directly on a multi-crystalline Si substrate for the first time. Several double-hetero (DH)-type p–n junctions were fabricated, and their optical emission properties were investigated.

II. EXPERIMENTAL

Multi-crystalline Si wafers with a thickness of 400 μm were used as substrates in this study. The surfaces were mirror-polished using a conventional Si wafer process. The wafers were highly p-type doped with a resistivity of ~1×10^{-2} Ω cm. The nanopillar crystals were grown on the substrates using a molecular beam epitaxy apparatus (UMB-300, Universal Systems) with two sets of conventional radio frequency (RF; 13.56 MHz)-excited nitrogen plasma cells. Purified N_2 gas was supplied to each plasma cell at 3 SCCM and excited at an RF power of 400 W. The substrate was thermally cleaned in a growth chamber at 965 °C for 10 min before growth. The morphologies of the samples were observed using field-emission scanning electron microscopy (FE-SEM, JSM-7800, JEOL). Current–voltage (I–V) characteristics were measured using the two-probe method, and electroluminescence (EL) spectra were measured using a spectrometer (C10082CA, Hamamatsu Photonics) at room temperature. The luminance of the light emission from the diodes was evaluated using a luminance meter (CS-160, Konica Minolta).

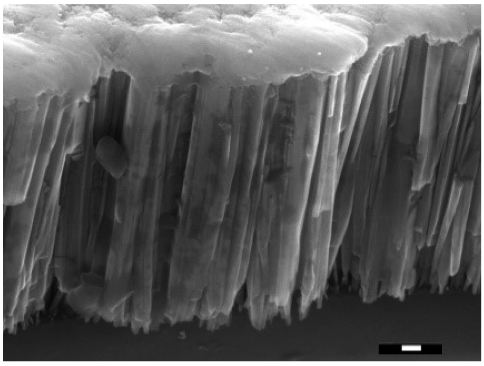

FIG. 1. Representative cross-sectional image of the prepared nanopillar-based diode after deposition of the ITO electrode. Scale bar indicates 100 nm.

III. RESULTS AND DISCUSSION

First, (In)GaN-based nanopillar crystals ~300 nm in height were directly grown on the substrate as steering crystals at 900 °C without nucleation layers between them, and they aligned in the direction perpendicular to the substrate surface. Surface problems, such as Ga melt-back etching, were not observed in this study. The crystals were aligned along the c axis, but were highly twisted to each other in the c plane.[26] During growth, gallium (7N) and indium (7N) were simultaneously supplied from individual effusion cells. The In composition of the steering crystals was ~1%. Generally, nanopillar crystals can be obtained under nitrogen-rich growth conditions, and the simultaneous supply of almost similar amounts of In with Ga and re-evaporation of most of the supplied In during growth were considered because such nanopillar crystals can be grown without nucleation layers.[21]

Germanium-doped n-type GaN and magnesium-doped p-type GaN nanopillar crystals were successively grown on steering crystals at a growth temperature of 900 °C. Their heights were approximately 600 nm, and their diameters ranged from 100 to 200 nm. For the DH-type junction, undoped InGaN nanopillar crystals ~150 nm in height were inserted between the n- and p-type crystals as the active region. Tin-doped indium oxide (ITO) thin films of 200 nm thickness were evaporated via electron-beam evaporation as ohmic electrodes to the Mg-doped GaN nanopillar crystals. The ITO electrodes were 1 mm φ in size and were annealed in an O_2 atmosphere at 500 °C for 5 min in an infrared furnace. A representative cross-sectional image of the prepared nanopillar-based diode is shown in Fig. 1. The nanopillar crystals aligned along the vertical direction of the substrate surface; however, the degree of alignment was not always as high as that of epitaxially grown crystals on single crystal substrates; for example, the full width at half maximum (FWHM) of the x-ray rocking curve for (0002) diffraction was ~500 arc min. The surface roughness of the layers composed of nanopillar crystals was evaluated to be approximately within 50 nm by SEM observation.[26]

The I–V characteristics, appearance of light emission, and EL spectrum of the prepared typical DH-type p–n-junction diode are shown in Fig. 2. For the growth of the n- and p-type GaN nanopillar crystals, the beam-equivalent pressure (BEP) of Ga was set to 1.9×10^{-6} Torr, and the dopant cell temperatures of Ge and Mg were set to 950 and 390 °C, respectively, whereas the growth temperature was set to 900 °C. Moreover, the BEPs of Ga and In were set to 5.4×10^{-7} and 7.2×10^{-8} Torr, respectively, and the growth temperature was reduced to 800 °C for the growth of the InGaN active region in the DH-structure diode. The I–V characteristics exhibited rectifying behavior, whereby the current increased at ~2 V. The highly p-type-doped multi-crystalline Si substrate can be utilized as an ohmic electrode for the (In)GaN steering crystals, which were directly grown on the substrate. The ohmic properties of the other components, except the DH junction regions, were confirmed; therefore, it was speculated that the rectifying behavior originated from the p–n-junction.

The measured luminance obtained from the DH junction was ~300 cd/m^2 at this time. The FWHM of the emission peak was ~40 nm. The diodes were directly grown on the multi-crystalline substrate; therefore, the performances were inferior to those of the LEDs grown on single-crystalline substrates; however, the results were similar to practical values. The In composition in the InGaN active region of the diode was estimated to be ~10%.[21] The light emission may have originated from the near-band edge transitions in the active region; however, a red shift in the emission peak occurred in this case. Therefore, the possibility of a Stokes shift due to compositional fluctuations in the InGaN active region or the effect of peculiar levels in the nanopillar crystals should be considered as contributing factors.

FIG. 2. *I–V* characteristic, photograph of light emission, and EL spectrum of the DH-type pn-junction diode directly grown on a mirror-polished multi-crystalline Si substrate.

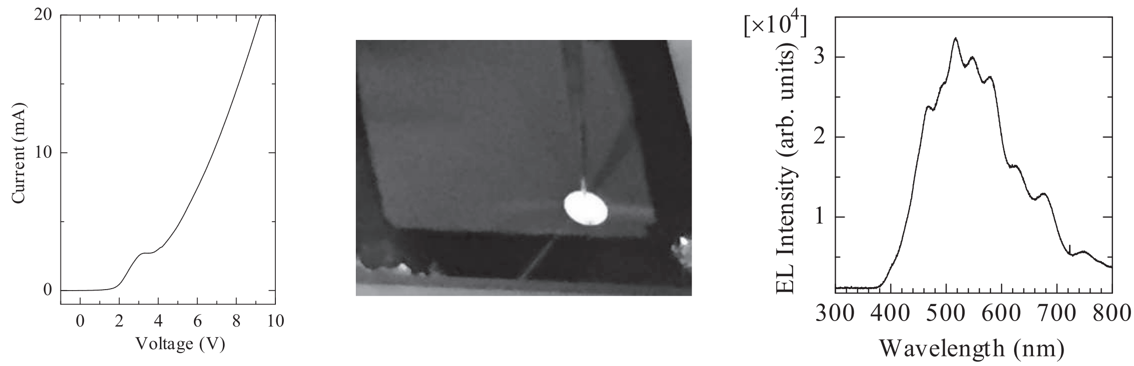

FIG. 3. *I–V* characteristic, photograph of light emission, and EL spectrum of the DH-type, white-light-emitting pn-junction diode directly grown on a multi-crystalline Si substrate.

In particular, white-light emission was observed for another DH-type diode, as shown in Fig. 3. The growth temperature of the InGaN active region was maintained at 800 °C, whereas the BEP of In decreased to 4.0 × 10^{-8} Torr in this case. A broad optical emission spectrum corresponding to the visible region was obtained, and a tunnel-diode characteristic was observed in the *I–V* curve. Blue–green light emission and white-light emission were obtained from the LEDs grown directly on a multi-crystalline Si substrate. Generally, the optimum growth temperature of InGaN alloys increases with the Ga composition.[27] In this study, the active layers in DH-LEDs were grown at the same growth temperature. White-light emission was obtained from the LED, in which the BEP of In was reduced compared with that of the LED that emitted blue–green light. In such a case, the Ga composition in the active layer increased; therefore, the optimum growth temperature increased with the Ga composition. In contrast, the growth temperatures of both the active layers remained constant. In other words, the growth temperature of the active layer in the LED that emitted white light was not sufficiently high for growth.

Reshchikov and Morkoç mentioned that point defects in undoped epitaxial GaN layers are related to broadband photoluminescence.[28] Broadband luminescence has been observed in several samples grown at relatively low temperatures. The emission peak was located at ~2.6 eV in their sample, and the FWHM was ~0.5 eV. The peak wavelength was closer to the blue wavelength; therefore, the color was aquamarine and labeled as the aquamarine luminescence (AL) band. The white light obtained in this study was assumed to be caused by defects similar to those that resulted in the AL band. The peak photon energy of the electroluminescence of the obtained In-included and nanopillar-based DH-LED grown on the multi-crystalline substrate in this study was located at ~2.4 eV (~515 nm) and was lower than that of the AL band. Differences in the compositions and hence the energy bandgaps among them are considered as the causes of the peak shift; more precise analyses are required in the near future to clarify these differences.

IV. CONCLUSION

In conclusion, several DH-type p–n-junction diodes of GaN-based nanopillar crystals were successfully and directly grown on multi-crystalline Si substrates for the first time. Although they were grown on a non-single crystalline substrate with no epitaxial relationships to group-III nitride crystals, clear light emissions were obtained. Therefore, multi-crystalline Si wafers can be an effective substrate for realizing novel, large-area, high-performance light-emitting devices.

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DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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