APPLIED PHYSICS LETTERS VOLUME 76, NUMBER 16 17 APRIL 2000

Polycrystalline nitride semiconductor light-emitting diodes fabricated on quartz substrates

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(Received 2 November 1999; accepted for publication 11 February 2000)

We demonstrate the feasibility of polycrystalline nitride semiconductor light-emitting diodes (LEDs). Here, polycrystalline LEDs were deposited on quartz substrates, incorporating a layer structure identical to that used for epitaxially grown LEDs. The deposition exhibits a tendency to produce *c*-oriented crystallites. Violet-blue (430 nm) operation of a polycrystalline LED is demonstrated, with spectral width of 38 nm, and emission efficiency approximately two orders of magnitude lower than for single-crystal LEDs. These LEDs could potentially be incorporated in large-area displays, since the deposition of polycrystalline materials avoids single-crystal substrates required for conventional nitride semiconductor light emitters. © *2000 American Institute of Physics.* [S0003-6951(00)02115-X]

Crystalline InGaN/AlGaN heterostructures, typically deposited epitaxially on sapphire or SiC substrates by organometallic vapor phase epitaxy (OMVPE), can be used for highly efficient visible light emitters.[1–4] For example, when combined with existing red emitters, the brightness and color purity of blue and green InGaN/AlGaN light-emitting diodes (LEDs) enable outdoor, full-color displays. The high efficiency of nitride LEDs is nevertheless very surprising and unusual compared to other compound semiconductor LEDs, because of the high density ($\sim 10^{10}\,\mathrm{cm}^{-2}$) of dislocations running through the material.[5] Apparently, these extended defects do not influence carrier flow and recombination in devices fabricated from crystalline material. If these defects are truly so inert, efficient LED operation may also be possible from even more highly dislocated material, for example randomly oriented polycrystals grown on glass substrates. In this letter, we demonstrate that polycrystalline nitride semiconductor LEDs can be produced on quartz substrates.

The polycrystalline nitride LED growth on quartz followed an OMVPE growth sequence identical to the procedure for growing single crystal films on sapphire substrates.[6] First, a thin (~ 30 nm) amorphous GaN buffer layer was deposited at 550 °C, and solid-phase crystallized by increasing the temperature to 1050 °C in an NH_3/H_2 ambient. The polycrystalline LED structure subsequently grown was very simple, including only 4 μm *n*-type GaN:Si, a 30 Å $In_{0.2}Ga_{0.8}N$ single quantum well, and a 0.2 μm *p*-type GaN:Mg layer. This structure is indicated in Fig. 1.

A single-crystal LED reference sample was simultaneously grown on a *c*-oriented sapphire substrate, for comparison of structural and performance characteristics. From these materials, crude LEDs were fabricated by evaporating Ti/Au *p*-contact metal, patterning the metal into $\sim 500\,\mu$m dots on 1 mm centers, and argon-ion milling the surrounding material. The etch depth was about 1 μm, as is typical for epitaxial LEDs grown on sapphire substrates, to expose the underlying *n*-type material. For LED operation, the *n*-type contact was made by simply touching a metal probe tip to the *n*-type GaN:Si surface (i.e., no *n* metal was deposited).

Unlike the specular surface obtained for single crystal LED material grown on sapphire substrates, the surface of the polycrystalline material grown over a quartz substrate was quite rough, as can be seen in Fig. 2, a scanning electron micrograph (SEM) of the polycrystals. Because of this irregular surface, the $p-n$ junction may not be removed uniformly by the ion milling. Furthermore, this surface morphology makes the nature of the *p*-semiconductor/metal contact ambiguous, since the relatively thin layer of *p*-type GaN:Mg may not completely coat each of the crystallites.

In the SEM image of the surface, the typical size of the crystals is a few μm, or roughly the same order as the film thickness. Furthermore, the presence of many flat-topped, hexagon-cross-section crystallites suggests that there is a tendency toward growth in a *c* orientation.[7] This preferred orientation is likewise evident in the x-ray diffraction spectrum, shown in Fig. 3, which contains a (002) reflection that is

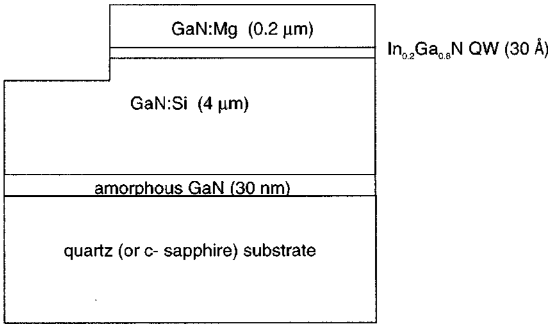

FIG. 1. Schematic diagram of nitride LED structure grown on quartz or sapphire substrates.

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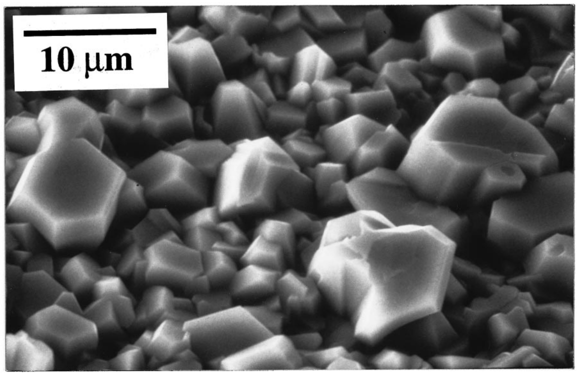

FIG. 2. Scanning electron micrograph of nitride LED polycrystals deposited on quartz substrates.

TABLE I. Measured x-ray diffraction intensities compared to documented values for a random arrangement of GaN polycrystals (JCPDS).[a] Both sets of intensities are normalized to the (101) diffracted intensity, which is the strongest reflection for perfectly random polycrystals.

Angle (2θ)	index	Measured $I/I_{(101)}$	JCPDS $I/I_{(101)}$
32.41°	(100)	0	70
34.56°	(002)	3521	50
36.82°	(101)	100	100
48.09°	(102)	116	60
57.91°	(110)	0	90
63.42°	(103)	258	80
67.75°	(200)	0	20
69.17°	(112)	21	80
70.60°	(201)	0	70
72.88°	(004)	116	20
78.38°	(202)	0	50

[a] Ref. 8.

strong compared to the intensity expected for an array of randomly orientated polycrystals, such as occur in powders. In Table I the diffracted intensities are compared with tabulated values corresponding to perfectly random polycrystals.[8] In each case, the intensities are normalized to the (101) reflection, because it is expected to be the strongest reflection from a random assembly of crystallites. This summary shows that the (002) reflection is especially pronounced, indicating a preferred *c* orientation for the nitride film grown on quartz. Accordingly, the (004) reflection is also relatively strong, and several of the asymmetric reflections are absent in the measured spectrum. The *c* orientation is also favored for single-crystal films grown on sapphire substrates, in which case *c*-oriented nitride films result for growth on either *c*-, *a*-, *r*-, or *m*-face Al_2O_3.[9]

Although the orientation of the polycrystals is not completely random, the structure is still highly disordered, such that many natural growth faces are still exposed during growth of the InGaN quantum well (QW). Since the growth rate and alloy composition are influenced by the plane upon which a layer is grown, it is natural to expect a range of QW parameters among the polycrystals, certainly broader than the distribution which occurs for single-crystal LEDs. Such a range of QW thickness and composition should translate into less spectral purity for the LED emission. Indeed, this may be evident in Fig. 4, a comparison of the emission collected through the substrate, of a single crystal LED (sapphire substrate) and a polycrystalline LED (quartz substrate) from the same growth run. While both spectra peak at ~430 nm, the spectral width of the polycrystalline LEDs is nearly double that of the single-crystal LEDs. However, it is important to note that it is not possible to verify the existence of the InGaN QW from this emission spectrum. The relatively broad spectral peak at 430 nm is also consistent with the emission resulting from recombination in magnesium-doped GaN.

Compared to the single-crystal LEDs, the detected emission is approximately 100 times less intense for the polycrystalline devices. Despite this relatively weak electroluminescence from the polycrystalline LEDs, several factors inhibit a meaningful comparison of the emission efficiency for the two kinds of LEDs. In particular, the unoptimized contact arrangement, along with a poorly defined current path in the polycrystalline LEDs may contribute to their low efficiency. For instance, the injection path is well confined in the single crystal LEDs, such that light is emitted only under the *p* contact. In contrast, the emission pattern from the polycrystals was much more complicated, with many bright emission spots observed in the region between the two contacts (and not only under the contact). This distributed emission was not effectively collected by our fiber-coupled spectrometer, thus contributing to the apparent inefficiency indicated by Fig. 4.

Such a scattered emission pattern may not be so unusual, however, for LEDs constructed from polycrystalline mate-

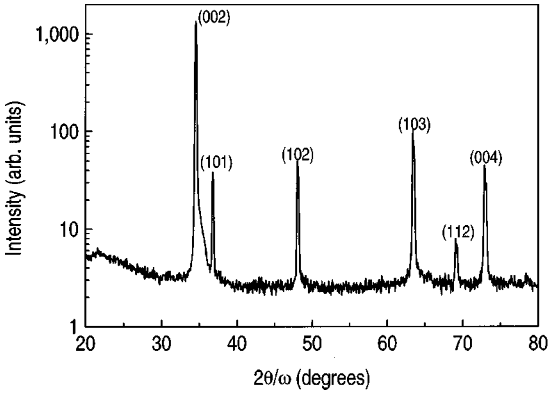

FIG. 3. X-ray diffraction spectrum of polycrystalline nitride LED structure grown on a quartz substrate.

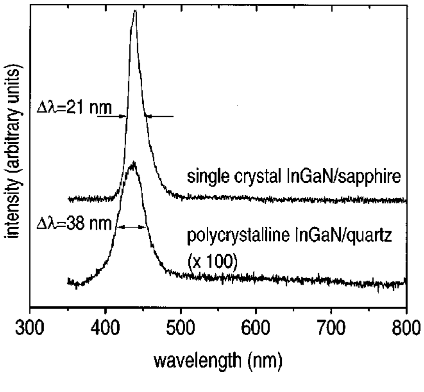

FIG. 4. Emission spectrum of nitride LEDs at 2 mA drive current: top-single crystal LED grown epitaxially on a *c*-face sapphire substrate; compared to bottom-polycrystalline LED deposited on quartz.

rial. The multitude of grain boundaries present in polycrystalline films forces the injected current to traverse a complicated path through the crystallites, in order to be injected across the junction. Consequently, when viewed with a microscope, the emission contains many small, bright spots, both between and around the two contacts, representing the percolation path assumed by the diode current. Further optimization of the device structure, for instance more sophisticated etching which would uniformly remove the junction, or further patterning of the material to define a current injection path, could potentially contribute to more efficient luminescence from these devices.

In conclusion, we have demonstrated the feasibility of polycrystalline nitride semiconductor LEDs, of potential utility for large-area displays, since polycrystalline materials can be deposited on much larger substrates than conventional single-crystal LED materials. Electroluminescence at 430 nm was obtained from a nitride LED, with emission efficiency approximately two orders of magnitude lower than for single-crystal LEDs grown on sapphire substrates. This structure was deposited on a quartz substrate, for which the nitride crystallites exhibited a tendency to be *c* oriented.

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