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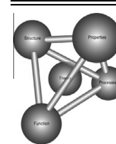

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Multilayered InGaN/GaN structure vs. single InGaN layer for solar cell applications: A comparative study

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Abstract

We report a comparison of the morphological, structural and optical properties of both InGaN single-layer and multilayered structures, the latter consisting of periodic thin GaN interlayers inserted during InGaN growth. It is shown that such a structure suppresses the In concentration fluctuations and corresponding different states of strain relaxation with depth, both detrimental to solar cell applications. Measurements performed by X-ray diffraction, cathodoluminescence and photoluminescence demonstrate that this multilayer growth is a promising approach to increase both the InGaN layer total thickness and In content in InGaN epilayers. As an example, single-phase 120 nm thick InGaN with 14.3% In content is obtained and found to possess high structural quality.
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1. Introduction

$In_xGa_{1-x}N$ material, with its direct bandgap covering continuously the full visible range of the solar spectrum by varying only the In content, enables the production of high-efficiency solar cells based solely on nitride material [1]. To obtain this high efficiency, InGaN layers with a large In content are required. For instance, a solar cell with only one InGaN junction containing 68% indium, can achieve 30% efficiency [2]. In addition, despite a high absorption coefficient, InGaN layer thicknesses >100 nm are required for the absorption of more than 90% of the incident above-bandgap light [3]. To date, the maximum In incorporation in InGaN-based solar cells with thicknesses >100 nm grown by metalorganic vapor-phase epitaxy (MOVPE) ranges from 12% to 25% [4–9]. At such high In contents, due to the low crystalline quality of the InGaN layer, bandgap energy fluctuations and carrier recombination at localized states arise. The conversion efficiency of such devices is thus limited, leading to low values of the open-circuit voltage and short-circuit current density [8,9]. Thus, it is challenging to investigate new InGaN layer growth techniques that allow high In incorporation in thick layers while maintaining high crystalline quality. The main issue arises from the broad immiscibility region between the two binary compounds InN and GaN, which originates from their large relative lattice mismatch, and leads to phase separations and/or In fluctuations with depth [10]. This has been observed in both thick InGaN layers [11–14] and in InGaN multi-quantum-well (MQW) structures

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[10,12,15]. Furthermore, literature data report that for layer thickness and In incorporation >100 nm and >10%, respectively, distinct sublayers exist in the different InGaN layers [16–19,11]. Close to the InGaN/GaN interface, the film is two-dimensional (2-D), homogeneous and fully strained on the GaN template substrate. As the growth proceeds, we have shown that spatial fluctuations of the composition are observed and an accumulation of In arises especially around threading dislocation terminations leading to 3-D In-rich domains embedded in the 2-D InGaN matrix [19,14]. Such local In-rich regions contribute to the transition from 2-D to 3-D growth observed in thick epilayers. Recently, we proposed growing InGaN epilayers by periodically inserting ultrathin GaN interlayers during the growth process to absorb the excess In and relieve the InGaN compressive strain [20]. This approach was shown to allow the growth of thick single-phase InGaN layers of high crystalline quality.

Thus, the aim of this paper is to compare in detail the properties of both InGaN/GaN multilayered (M-sample) and InGaN single-layer (C-sample) structures, grown with the same total InGaN layer thickness and nominal In composition. In particular, optical emission properties are investigated using room-temperature depth-resolved cathodoluminescence (CL) and temperature-dependent photoluminescence (PL). It is found that the multilayered approach effectively suppresses the distinct sublayers and enhances the InGaN material quality as compared to the conventional growth of a thick single-layer structure. Using this approach, we succeed in obtaining single-phase 120 nm thick InGaN containing 14.3% In that exhibited high structural quality.

2. Experiment

Two set of InGaN samples were grown at 800 °C on GaN templates in a T-shape MOVPE reactor [21]. Nitrogen (N_2) was used as the carrier gas and trimethylgallium (TMGa), trimethylindium (TMIn) and ammonia (NH_3) were employed as precursor sources for Ga, In and elemental N, respectively. The reactor pressure was 100 Torr and the V/III ratio equal to 8000. The ratio of TMIn to the sum of TMIn and TMGa in the vapor phase (TMIn/III) was kept constant at 12.5% for the first set of samples, and then increased to 45% for the second set of samples. For each set of samples, a reference InGaN single-layer (C-sample) and a multilayered structure (M-sample) were grown for comparison. A schematic representation of both structures under study is shown in Fig. 1. The M-samples were grown by periodically stopping the In precursor flow into the reactor. The two targeted structures consist of 5×1.5 nm (first set of samples) and 16×1.5 nm (second set of samples) thick GaN interlayers, inserted periodically between 6×21 nm (first set of samples) and 17×7 nm (second set of samples) thick InGaN layers, respectively, resulting in a total structure thickness of 133.5 nm (first set of samples) and 143 nm (second set of samples). The InGaN and GaN periodicity and layer thicknesses are based on simulations and previous experimental results on the relaxation of InGaN layers [19]. The layer thicknesses are deduced from in situ reflectometry and confirmed by (00.2) $\omega - 2\theta$ X-ray diffraction measurements by applying a fitting model to the Penddelosung fringes. Symmetric (00.4) $\omega - 2\theta$ scans in combination with X-ray reciprocal space maps (RSMs) of the asymmetric (10.5) planes are used to determine the lattice parameters (a, c), the degree of relaxation and the In content in the different samples. The input equations and parameters necessary to obtain these data are taken from Ref. [22]. The isocomposition lines are calculated according to the model proposed by Pereira et al. [23]. The surface morphology is observed by scanning electron microscopy (SEM) and atomic force microscopy (AFM). Microcompositional studies through energy dispersive X-ray spectroscopy (EDX) are carried out in an aberration-corrected JEOL 220FS microscope, operating at 200 kV with a probe current of 150 pA, and a probe size of 0.12 nm at the full width at half maximum (FWHM). Optical emission properties of InGaN structures are investigated by both CL

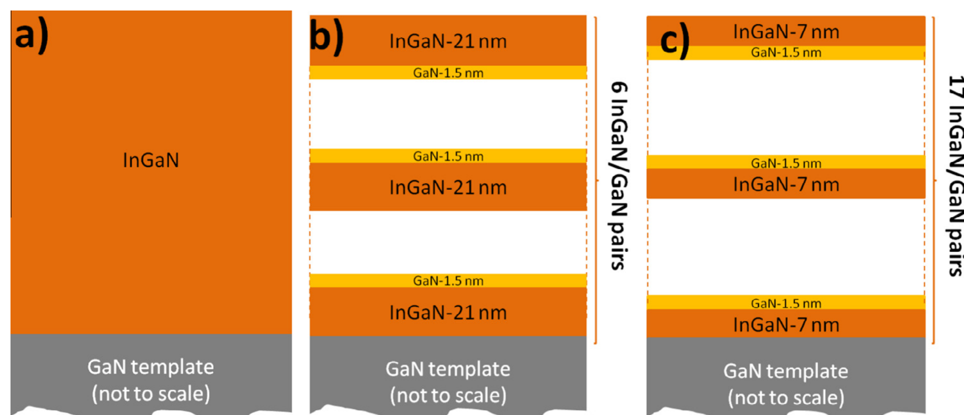

Fig. 1. Schematic representation of (a) C-sample, (b) M-sample of the first set of samples and (c) M-sample of the second set of sample structures. The InGaN total thickness and the nominal In composition are equal to 126 nm and 120 nm, and 10.2% and 14.3% for each set of samples, respectively.

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