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Phase separation suppression in $In_xGa_{1-x}N$ on a Si substrate using an indium modulation technique

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ABSTRACT

A high quality, single phase InGaN film is fabricated on a GaN/Si (111) substrate by optimizing the pulse patterned In supply with a plasma-assisted MBE technique. Compositional phase separation in InGaN is considerably suppressed. The optical and structural properties of the single phase InGaN epitaxial film are consistently confirmed by atomic force microscopy, X-ray diffraction and photoluminescence measurements. We propose a growth mechanism for single phase InGaN in terms of optimal incorporation and surface migration of In atoms.

1. Introduction

$In_xGa_{1-x}N$ alloys play a dominant role in the field of blue and green solid state lighting technology due to their direct and tunable band gap covering the entire visible and near infrared range. This is done by varying the In composition from $x = 1$ (0.7 eV) to $x = 0$ (3.4 eV) [1]. However, the quantum efficiency of $In_xGa_{1-x}N$ becomes quickly degraded with increasing In composition (>0.2) [2]. Growth of high quality $In_xGa_{1-x}N$ with high x (>0.2) is essential to develop high performance optoelectronic devices for long wavelength (λ) light sources (λ > 530 nm), as well as for high efficiency photovoltaic solar cells [3–5].

Growing nitride materials on hetero substrates with different lattice constants is challenging due to the large lattice mismatch at the heterojunction. This unavoidable lattice mismatch leads to crystalline defects in the nitride film. Examples are inversion domain boundaries and stacking faults that generate nonradiative recombination states in the band gap [6]. Due to their small lattice mismatch, sapphire and SiC are widely and successfully used as a substrate. Si is considered to be the most suitable alternative due to its cost effectiveness and large area InGaN growth [7]. Another core issue is developing a method to precisely control In composition of InGaN, especially for InGaN with a high In content. In order to grow InGaN with a high In content, a low growth temperature is generally used to avoid In–N thermal decomposition [8]. However, this low temperature growth causes nonuniform local three dimensional growth due to the short migration length of constituting atoms on the surface [9,10]. Although migration enhanced epitaxy under metal-rich conditions has been proposed to overcome this problem, excess metal atoms at the growth front often leads to metal segregation resulting in metal-rich localized states or metal clustering [11]. Therefore, appropriate control of III-V stoichiometric and thermal conditions is necessary to fabricate high quality InGaN [8,12,13]. The third issue is to minimize phase separation of InGaN that is linked to the miscibility gap between InN and GaN [14,15]. Misfit strain accumulation caused by the phase separation during InGaN growth plays an important role in determining the film's structural and compositional

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qualities [16]. Therefore, it is necessary to suppress the phase separation.

In this work, we developed a metal modulation epitaxy combined with plasma-assisted MBE to suppress the phase separation in InGaN grown on a Si (111) substrate. The degree of the phase separation in InGaN was investigated by measuring photoluminescence (PL), and the single phase PL spectrum was observed in the optimized InGaN film. We propose a mechanism to minimize the phase separation in InGaN prepared by an In modulation epitaxy.

2. Experimental

InGaN epitaxial growth on a 2-inch Si (111) substrate was carried out in a MBE system, using a standard effusion cell for group III sources. A radio frequency (RF) plasma source was employed for active nitrogen (N^*). The native oxide layer on the Si substrate was removed by the thermal dissociation process after depositing a thin Ga layer. In this process, the gallium oxide was formed by the reaction between silicon oxide and Ga. This was subsequently thermally dissolved at 750 °C [17]. An oxide-free Si surface was confirmed by reflection high energy electron diffraction (RHEED). The GaN on Si template consisted of a 200 nm thick AlN layer, compositionally graded 50 nm thick AlGaN transition layers, and a 1 μm thick GaN buffer layer. Finally, a 100 nm thick InGaN layer was grown at 550 °C of substrate temperature with a pulse patterned In supply. The source flux of constituting Ga and In atoms were $F(\mathrm{Ga}) = 2.7$ ML/min and $F(\mathrm{In}) = 2.5$ ML/min, respectively. Active nitrogen was supplied at a rate of 1.0 sccm with RF plasma power at 200 W. This corresponds to $F(\mathrm{N}^*) = 3.5$ ML/min of beam flux. Fig. 1(a) shows the schematic diagram of the In modulation epitaxy. The Ga and N^* fluxes were continuously supplied during the growth, while the In flux was periodically interrupted. In supply time was varied from 9 to 30 s, while the interruption time was kept to 10 s for all the samples (refer to Table 1). These two regimes were periodically iterated. InGaN growth was monitored in situ by RHEED. The structural properties and surface morphologies of the samples were characterized by X-ray diffraction (XRD) 2θ−ω scans and atomic force microscopy (AFM). PL measurements were performed with a 266 nm DPSS laser source at 10 K.

3. Results and discussion

Fig. 1(b) shows the growth time dependent RHEED intensity during the In modulation epitaxy with a period of 21 s In supply time. The insets A and B show the RHEED patterns at the minimum and maximum RHEED intensities respectively. The intensity is periodically oscillated when the amount of the metallic elements (Ga and In) on the surface is changed, and this corresponds to the cycle of In supply. The spike-like noise is caused by substrate rotation. Since the Ga–N binding energy is larger than the In–N binding energy, an In-rich surface is generally favored and therefore the metal adlayer is expected to mainly consist of In. At the end of the In supply regime ($F(\mathrm{Ga}) + F(\mathrm{In}) > F(\mathrm{N}^*)$), the metal adlayer becomes a maximum amount and the RHEED intensity reaches the minimum level due to the metal diffraction. In the interruption regime ($F(\mathrm{Ga}) < F(\mathrm{N}^*)$), the accumulated In atoms participate to produce InGaN nucleation, or are partly vaporized. As the amount of In adlayer approaches the minimum level, the RHEED exhibits a maximum with a clear (1 × 1) reconstruction (right inset). Therefore, the intensity of the RHEED pattern oscillates according to the period of the In supply.

However, different RHEED characteristics were observed during the growth with shorter and longer In supply time. For short In supply time (9 and 15 s), as the In supply depletes, a spotty pattern

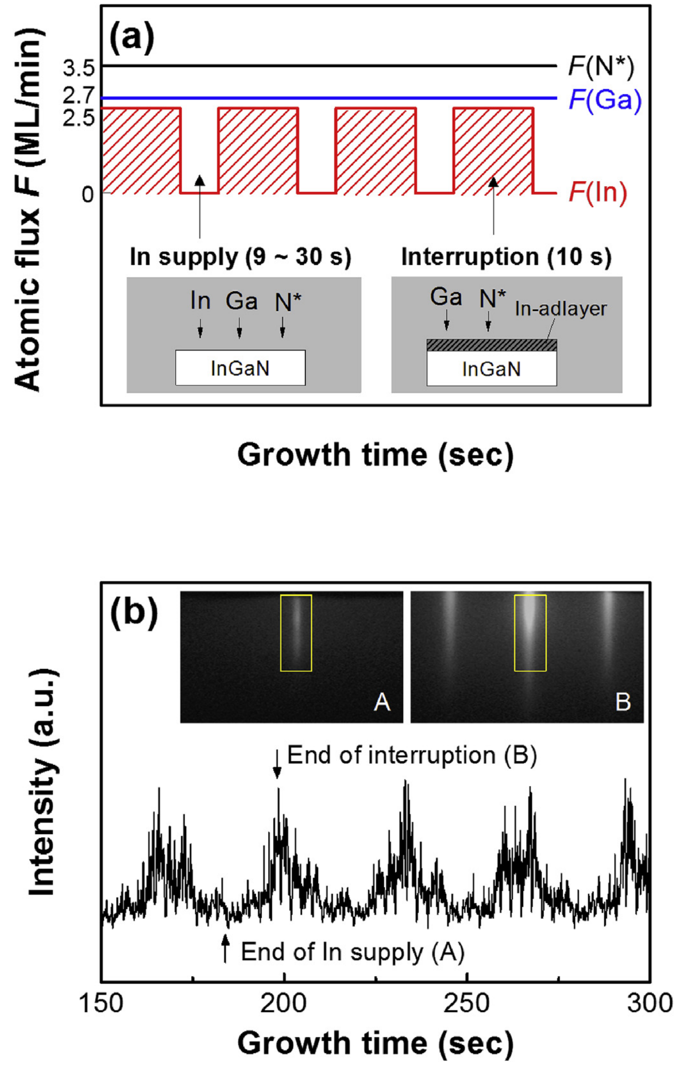

Fig. 1. (a) Schematic diagram explaining atomic fluxes (*F*) versus growth time. The In flux *F*(In) is modulated. The inset describes a proposed growth model for the epitaxial InGaN films. (b) Growth time dependent RHEED intensity during the modulation epitaxy (In supply time: 21 s and interruption time: 10 s). The insets A and B show the RHEED patterns which correspond to the minimum and maximum intensities respectively.

was revealed after exhaustion of the In adlayer. This indicates that the surface morphology becomes rough by three-dimensional growth under the N-rich condition. On the other hand, for the growth with long In supply (30 s), a minimum level of the intensity was maintained during the entire growth. The growth front becomes extremely In-rich due to the insufficient interrupt time of 10 s. In this case, the In droplets on the surface were observed by

Table 1
Growth parameters for InGaN epitaxy.

Parameter	Type of sample			
	a	b	c	d
In supply time (sec)	9	15	21	30
III/V flux ratio per cycle	1.11	1.20	1.26	1.31
In/Ga flux ratio per cycle	0.44	0.56	0.63	0.69
Maximum In adlayer (ML)	0.20	0.33	0.46	0.65
In composition from XRD	0.07	0.10	0.13	0.15
In incorporation efficiency, η	0.23	0.28	0.33	0.37