Effects of pulse cycle number on the quality of pulsed atomic-layer epitaxy AlN films grown via metal organic chemical vapor deposition

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The effect of different numbers of Al and N$_2$ pulse cycles on the quality of AlN films prepared via the pulsed atomic-layer epitaxy (PALE) technique and epitaxially deposited on a c-plane sapphire substrate by metal organic chemical vapor deposition were investigated. The characteristics of AlN/sapphire were studied by atomic force microscopy, field emission scanning electron microscopy and Raman spectroscopy. AlN film deposited with the PALE technique with the highest number of pulse cycles (1050) was observed to exhibit highly uniform spherical AlN grains and a dense film surface with a root mean square roughness of 0.46 nm. Transition of the E$_2$ (high) peak from the Raman spectrum shows that the strain compression in PALE AlN films is inversely proportional to the number of pulse cycles.

1. Introduction

AlN epilayers with a high crystal quality and low threading dislocation on foreign substrates have become essential for numerous optoelectronic and power device applications due to their large intrinsic energy bandgap and excellent miscibility with other III-nitride elements, allowing the formation of a variety of ternary and quaternary materials with a tunable lattice constant and energy bandgap.\(^1\)–\(^3\) Additionally, wurtzite AlN crystals have been shown to be promising candidates for piezoelectric sensors and actuators due to their large intrinsic polarization.\(^4\)–\(^5\)

Recently, metal organic chemical vapor deposition (MOCVD) has gained attention for the large-scale manufacturing of nitride thin films; their growth rates are relatively high and ternary and quaternary materials can be grown simultaneously in one growth cycle.\(^6\),\(^7\) Typically, AlN thin films are grown at high temperatures, but it is challenging to grow ternary and quaternary group III elements containing Al alloys at such temperatures due to the difference in the growth window for different elements.\(^8\),\(^9\) Therefore, it is preferable to produce AlN epilayers at comparatively low growth temperatures while maintaining the crystal quality. However, growth at low temperatures is characterized by a high number of parasitic gas phase pre-reactions between trimethylaluminum (TMAI) and NH$_3$, which decreases the growth rate and degrades the properties of the epilayer.\(^10\),\(^11\)

Recently, the pulsed atomic-layer epitaxy (PALE) technique has been reported as one of the most promising methods to overcome this issue. This technique involves supplying Al and N$_2$ alternately to the reactor, thus reducing the parasitic gas phase pre-reactions between these two elements. As a result, high-quality epilayers with low threading dislocation densities (TDDs) can be obtained at 100–200°C below typical AlN growth temperatures for conventional MOCVD.\(^12\),\(^13\) However, there has yet to be a systematic study on the effect of pulse cycle repetitions on the crystal quality of deposited AlN films via the PALE technique.

In this study, a crack-free, dense and smooth AlN epilayer deposited on a c-plane sapphire substrate via MOCVD at a relatively low growth temperature of 1180°C was achieved by sandwiching a thin AlN nucleation layer grown using nominal growth technique between the c-plane sapphire substrates and an AlN epilayer grown via the PALE technique. The influence of film thickness on the quality of deposited top PALE AlN films was investigated by varying the number of pulse cycles (350, 700 or 1050). The effect of the number of pulse cycles on the surface properties, morphological properties and structural quality of the deposited top AlN films is discussed in detail.

2. Experimental procedure

In this study, 2 inch (0 0 0 1)-sapphire substrates were employed. The epitaxial process was performed in a laminar MOCVD reactor with a continuous flow of H$_2$ gas as the carrier gas while N$_2$ gas was used as a sub-flow gas to initiate the growth process on the sapphire substrate. The N$_2$ and Al reactant source elements for the epitaxial growth were obtained from gaseous NH$_3$ and TMAI metal organic precursors.

First, the substrate was thermally cleaned at 1000°C for 10 min in flowing H$_2$ gas to eliminate the native oxide layer on the substrate surface without deteriorating the structure.\(^14\),\(^15\) Then, a 30-nm-thick AlN nucleation layer (NL) was grown by simultaneously flowing the TMAI and NH$_3$ source at 1100°C.\(^16\) Subsequently, an AlN film was deposited on top of the AlN NL by a NH$_3$ pulsed-flow growth method at a temperature of 1180°C. The pulse...