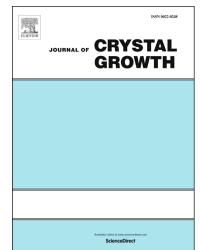
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ACCEPTED MANUSCRIPT

Effects of Deposition Temperature and Ammonia Flow on Metal-Organic Chemical Vapor Deposition of Hexagonal Boron Nitride

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Abstract

The use of metal-organic chemical vapor deposition at high temperature is investigated as a means to produce epitaxial hexagonal boron nitride (hBN) at the wafer scale. Several categories of hBN films were found to exist based upon precursor flows and deposition temperature. Low, intermediate, and high NH₃ flow regimes were found to lead to fundamentally different deposition behaviors. The low NH₃ flow regimes yielded discolored films of boron sub-nitride. The intermediate NH₃ flow regime yielded stoichiometric films that could be deposited as thick films. The high NH₃ flow regime yielded self-limited deposition with thicknesses limited to a few mono-layers. A Langmuir-Hinshelwood mechanism is proposed to explain the onset of self-limited behavior for the high NH₃ flow regimes could be further divided into low and high temperature behaviors with a boundary at 1500 °C. Films deposited with both high NH₃ flow and high temperature exhibited room temperature free exciton emission at 210 nm and 215.9 nm.

Keywords

A1. Adsorption; A2. Surface processes; A3. Chemical vapor deposition processes; A3. Metalorganic chemical vapor deposition; A3. Organometallic vapor phase epitaxy; B1. Nitrides

Introduction

Hexagonal boron nitride (hBN) is a wide bandgap (~6 eV) semiconductor of interest for use in optoelectronic devices, 2D heterostructures, as well as the more traditional uses in chemically and thermally stable coatings and components. The utility of hBN has been demonstrated in optoelectronics as release layers for III-nitrides on foreign substrates, thermal neutron detectors, and UV emitters ¹⁻⁵. hBN has also been shown to be an excellent substrate for graphene with hBN/graphene heterostructures exhibiting mobility of ~10⁵ cm² V⁻¹ s⁻¹, approaching the mobility of suspended graphene ⁶⁻⁸. However, high performance hBN based 2D heterostructures currently utilize hBN exfoliated from hBN single crystals a few millimeters in size, which limits scalability.

Prior studies of metal-organic chemical vapor deposition (MOCVD) of hBN using NH_3 and triethylborane (TEB) as precursor chemicals noted a number of interesting phenomena. Nakamura ⁹ showed that films deposited with a low V/III ratio presented a number of different hues. Compositional analysis showed that this discoloration was related to excess B; films deposited with a V/III ratio of 30 became B-rich at deposition temperatures below ~1000 °C and films deposited with a V/III ratio of 10 were B-rich over the entire experimental temperature range: 800 °C to 1200 °C. Kobayashi and Makimoto ¹⁰ observed

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