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Atomic ordering in $Al_xGa_{1-x}N$ thin-films

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Abstract

Samples of $Al_xGa_{1-x}N$ with $x_{Al} = 25\%$, 48%, and 78% were grown by metal organic chemical vapor deposition and examined using cross-sectional transmission electron microscopy. All samples were shown to exhibit both (0001) type ordering spots and streaking. The morphology of the ordered regions was determined to be plate-like on (0001) planes, with unequal axes in-plane.

Keywords: Single crystal growth; CVD; STEM; Compound semiconductors; Ordering

1. Introduction

$Al_xGa_{1-x}N$ thin-films are important semiconductors due to their large direct band gaps, which are dependent on the value of x. Such large direct band gaps have various useful consequences. The alloy has emission energies from 3.4 eV for pure GaN to 6.2 eV for pure AlN, this range results in light generation from shallow to deep ultraviolet (UV), allowing many novel optical applications. In addition to these applications, the formation of a two-dimensional electron gas at $GaN/Al_xGa_{1-x}N$ interfaces allows the fabrication of high electron mobility transistors [1]. These properties have led to great interest in the future production of UV detectors, short-wavelength lasers, and high-power white light-emitting diodes, which could provide more energy efficient, longer-lasting lights for use in everyday applications. The uses for the material have motivated research into fabricating $Al_xGa_{1-x}N$ with desirable electronic properties and crystalline quality. To produce the desired properties for any given application, the overall behavior of the system during growth must be understood.

$Al_xGa_{1-x}N$ alloys are known to order spontaneously during growth [2–5]. Various ordered structures have recently been discovered in this system. As in other III–V systems, such as the zinc-blende arsenides and phosphides, ordering occurs on the close-packed planes [6]. The only microstructural difference between the two structures is the stacking order of close-packed planes. The zinc-blende system exhibits three-layer repeating AaBbCcAaBbCc stacking, while the wurtzitic system exhibits two-layer repeating AaBbAaBb stacking. Unlike the zinc-blende III–V system though, the effects of the formation of ordered structures on the electronic properties of the III–N materials have not yet been studied extensively.

Various ordering ratios have been observed in the $Al_xGa_{1-x}N$ system [5]. An accepted candidate mechanism has not been advanced to explain the occurrence of a variety of ordering structures occurring within the system. Northrup et al. have proposed a mechanism of ordering for $In_xGa_{1-x}N$ materials to explain the occurrence of 1:1 ordered structures [7]. While this mechanism was not proposed for $Al_xGa_{1-x}N$, the systems are related, so their ordering mechanisms may be similar. This method involves preferential occupation of metal sublattice sites on $\langle 10\bar{1}1 \rangle$ facets due to preferential attachment of aluminum and gallium at certain positions in the lattice. This occurrence would yield 1:1 ordering in the [0001] direction. It would not though explain the other types of ordering seen in this system, although a full examination of energetically favorable atomic sites on the typical facet planes for $Al_xGa_{1-x}N$ has not been undertaken.

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In this study, the formation of ordered regions and their morphology in thin-film $Al_xGa_{1-x}N$ were studied using transmission electron microscopy (TEM) weak-beam dark-field (WBDF) imaging and selected area electron diffraction (SAED). Characteristics of the observed electron diffraction patterns from the samples are rationalized in terms of their underlying ordering structures and morphologies and are correlated with the observed WBDF micrographs. Calculations are presented to verify the structures represented by the diffraction patterns. The SAED patterns are also interpreted to provide insight into the morphology of the ordered domains observed. These results constitute the present paper.

2. Experimental

Thin-film $Al_xGa_{1-x}N$ samples with $x_{Al} = 25\%$, 48%, and 78% were grown using an AIXTRON 200/RF horizontal metal organic chemical vapor deposition (MOCVD) reactor. Temperatures were monitored by a thermocouple calibrated to the wafer temperature, and all temperatures given are in terms of the thermocouple set points. (0001) oriented sapphire wafers were used as substrates. These were not chemically cleaned prior to use, but were supplied in an epi-ready state and opened under a nitrogen atmosphere. The growth process began with a low-temperature nucleation layer of AlN on (0001) oriented sapphire at 560 °C. This was followed by an AlN buffer layer of 100–200 nm thickness grown at 1190 °C. This layer was intended to cause a compressive strain state in the subsequent $Al_xGa_{1-x}N$ layer and prevent crack formation. The $Al_{.25}Ga_{.75}N$ layer was then grown at 1170 °C and the two films with higher aluminum contents were grown at 1190 °C. The layers were grown using hydrogen as a carrier gas, an ammonia flow of 2 l/min, and a total group-III flow (TMGa + TMAl) of 30 sccm.

The composition of each sample was determined using Rutherford backscattering (RBS). A Bede diffractometer was then used for a preliminary examination of the $Al_xGa_{1-x}N$ layers to determine that the film was of good crystal quality before the wafers were sectioned to make TEM samples. Finally, electron microscopy was performed with a JEOL 4000-EX electron microscope with a LaB_6 filament operating at a 400 keV accelerating voltage. The images and SAED patterns were recorded on electron-sensitive film.

All TEM samples were prepared using the standard method of grinding, dimpling, and ion-milling to provide an electron-transparent region. The samples to be examined with TEM for the presence of ordering were prepared with a $[1\,0\,\bar{1}\,0]$ surface normal. The $[1\,0\,\bar{1}\,0]$ orientation prevents the formation of double diffraction spots, a dynamic effect that can mimic the diffraction pattern of an ordered structure. TEM samples with a surface normal of $[1\,1\,\bar{2}\,0]$ were also prepared for evaluating the morphologies of ordered domains.

3. Results

As the orientation for all TEM samples used for WBDF was $[1\,0\,\bar{1}\,0]$, the mutually normal reflections $\mathbf{g} = (0\,0\,0\,2)$ and $\mathbf{g} = (\bar{1}\,2\,\bar{1}\,0)$ were used to generate WBDF micrographs. Use of both these reflections allows observation and characterization of **a**-type, **c**-type, and **c** + **a** dislocations, as can be seen from Fig. 1(a)–(d). In Fig. 1(a)–(c), the only dislocations visible are **a** and (**c** + **a**)-type, while Fig. 1(d) shows **c**-type and (**c** + **a**)-type dislocations.

The vertical bands of light and dark contrast that obscure many of the dislocations in Fig. 1(a) and (b) are caused by formation of tilted regions in the sample. Such regions form as a result of the high initial strain state of the film. As can be seen from the TEM samples, the area near the interface is most defected, with the structure stabilizing as the film grows thicker. As the material relaxes,

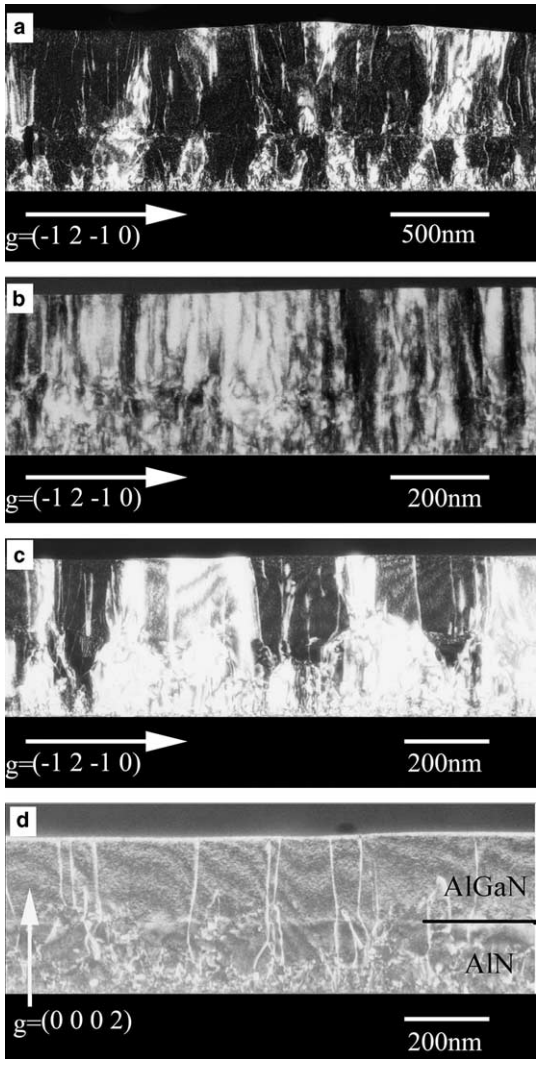

Fig. 1. Weak-beam dark-field images of cross-sectional samples obtained from (a) $x_{Al} = 25\%$, (b) $x_{Al} = 48\%$, (c) $x_{Al} = 78\%$ and (d) $x_{Al} = 78\%$ with the AlGaN layer, AlN layer, and interface marked. $\mathbf{g} = (\bar{1}\,2\,\bar{1}\,0)$ was used for (a), (b), and (c), while $\mathbf{g} = (0\,0\,0\,2)$ was used for (d). It should be noted that (c) and (d) do not represent the same area of the sample.