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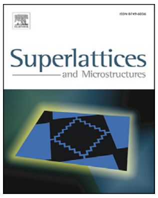

Residual stress in AlN films grown on sapphire substrates by molecular beam epitaxy

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ABSTRACT

Residual stress in AlN films grown by molecular beam epitaxy (MBE) has been studied by Raman scattering spectroscopy. A strain-free Raman frequency and a biaxial stress coefficient for E_2(high) mode are experimentally determined to be 657.8 ± 0.3 cm^{-1} and $2.4 \pm 0.2\ cm^{-1}/GPa$, respectively. By using these parameters, the residual stress of a series of AlN layers grown under different buffer layer conditions has been investigated. The residual compressive stress is found to be obviously decreased by increasing the Al/N beam flux ratio of the buffer layer, indicating the generation of tensile stress due to stronger coalescence of AlN grains, as also confirmed by the *in-situ* reflection high energy electron diffraction (RHEED) monitoring observation. The stronger coalescence does lead to improved quality of AlN films as expected.

1. Introduction

Recently, AlGaN-based quantum devices have attracted much research attention in the areas of electrical engineering, material science, and physics due to the numerous potential applications such as ultraviolet solid-state light sources, solar-blind photodetectors and so on [1,2]. Among the III-nitride semiconductors, AlN is always regarded as a preferred device substrate or template material for crack-free fabrication. Up to now, great progress has been made for the growth of either bulk AlN or AlN templates [3–6]. However, the device performance is still greatly limited by the strain-induced bending or cracks in thick AlN films commonly grown by hetero-epitaxy on sapphire, Si or SiC substrates [7,8]. In this sense, residual stress analysis in layered structures is of great importance for understanding the mechanical properties of AlN films and helps to adjust the strain status for crystalline quality improvement. Among all the techniques, Raman scattering spectroscopy provides a contactless, non-destructive and sensitive tool to characterize the stress distribution, making it an essential and potential way for residual stress analysis [9]. Generally, the biaxial stress distribution can be easily acquired by the linear

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dependence of biaxial stress on E_2(high) phonon frequency using the obtained strain-free Raman frequency and biaxial stress coefficient. Some efforts have been performed on this issue and both theoretical and experimental results have been explored [10–13]. However, most reports focused on MOCVD-grown or HVPE-grown templates which somehow disagreed with the theoretical values and the contradictions were not well explained. Few reports are available to precisely determine the above parameters from AlN films grown by molecular beam epitaxy (MBE).

MBE supplies us a way to obtain atomically flat and high-purity AlN templates. The avoidance of oxygen contamination is of great importance towards the high-quality AlGaN quantum wells [14,15]. In the early work, we have obtained a much broad growth window for droplet-free AlN films with step-flow feature [16]. But the crystalline quality can still be further improved through studying the strain states in the epi-layers. In this work, the crystalline quality of MBE-grown AlN films was improved by increasing the Al/N beam flux ratio during AlN buffer layer growth. Residual stress analysis showed that this improvement originated from a stronger AlN grains coalescence for higher Al/N ratio. This analysis is based on the obtained values of the biaxial stress coefficient and strain-free Raman frequency for E_2(high) mode, which have been experimentally determined in this work and found to be well in accord with the theoretically predicted values.

2. Experimental

The growth of AlN films was performed on the 2-inch c-plane sapphire substrates by plasma-assisted MBE system (SVTA). Reflection high energy electron diffraction (RHEED) and an optical reflection spectrometer are used to *in-situ* monitor the whole growth process. Sapphire substrate was first thermally cleaned at 1000 °C for an hour. Then, nitridation was performed at 800 °C for half an hour, leading to the formation of very thin AlN layer on the surface. AlN layers were then deposited by using low temperature AlN buffer layer. To evaluate the residual strain, tens of samples were selected at different growth conditions such as varied growth temperature, Al/N ratio and thickness as ever reported [16]. To study the effect of buffer layer growth condition on quality improvement, five samples with common structure of 30-nm-thick AlN buffer layers and 360-nm-thick AlN top layers were grown. Another sample without buffer layer, as a reference, was also grown. All the growth process is kept the same except that the Al/N ratio changes monotonically during the buffer layer growth. The lattice constants of the AlN films have been determined by high-resolution X-ray diffraction (XRD) measurements using $K\alpha_1$ ($\lambda = 0.15$ nm) radiation. Residual stress was estimated by Raman spectroscopy at room temperature in z(x, x)-z backscattering geometry using a laser excitation source at a wavelength of 514 nm.

3. Results and discussion

To study the biaxial residual stress in hexagonal AlN by Raman scattering measurement, one usually pays attention to E_2(high) mode since it corresponds to atomic oscillations in the c-plane [17]. Two parameters are important to study stress, i.e. strain-free Raman frequency for E_2(high) mode and the Raman biaxial stress coefficient (k). The in-plane residual stress (σ_a) can be derived by using $\sigma_a = k^{-1}\Delta\omega[E_2(high)]$, where $\Delta\omega[E_2(high)]$ is the strain-induced Raman frequency shift for E_2(high) mode. To determine the biaxial stress coefficient as accurate as possible, Raman scattering measurements were performed on a large number of samples grown on sapphire substrates by MBE. Fig. 1(a) shows a typical Raman scattering spectrum for AlN layer grown on the sapphire substrate, where several scattering peaks are observed. The peaks located at 657 and 890 cm^{-1}

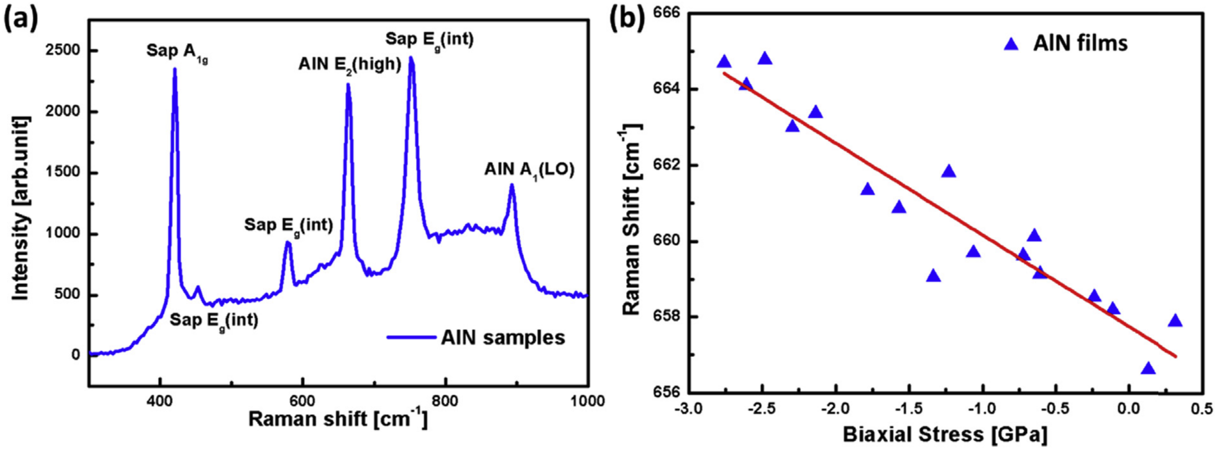

Fig. 1. (a) A typical Raman spectrum of MBE-grown AlN films on sapphire. Visible are the E_2(high) and A_1 (LO) modes in AlN films as well as A_{1g} and E_g(int) modes in sapphire substrate. (b) Raman frequencies of E_2(high) phonon mode as a function of residual biaxial stress. The red line shows a linear fit to the data, which gives a strain-free phonon frequency of 657.8 ± 0.3 cm^{-1} and a biaxial stress coefficient of 2.4 ± 0.2 $\boldsymbol{cm^{-1}/GPa}$ for the E_2(high) mode. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)