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Correlation between Raman intensity of the N-related local vibrational mode and N content in GaAsN strained layers grown by MOVPE

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Abstract

$GaAs_{1-x}N_x$ alloy films ($0 \leqslant x \leqslant 0.055$) grown on GaAs (0 0 1) substrates by metalorganic vapor phase epitaxy (MOVPE) using TBAs and DMHy as As and N precursors, respectively, have been investigated by Raman spectroscopy. It was found that, with incorporating N up to $x = 0.055$, a single N-related localized vibrational mode (LVM) is observed at around 468–475 cm^{-1}. We have investigated the N-related LVM Raman intensity (I_{LVM}) and frequency (ω_{LVM}) as a function of N concentration. Both the I_{LVM} and the ω_{LVM} were found to rise for the $GaAs_{1-x}N_x$ films with higher N incorporation. It is also evident that the N concentration in the $GaAs_{1-x}N_x$ grown films determined by Raman spectroscopy technique (x_{Raman}) exhibits a linear dependence on the N concentrations determined by the high resolution X-ray diffraction (HRXRD) (x_{XRD}). Our results demonstrate that the linear dependence of the x_{Raman} on the x_{XRD} provides a useful calibration method to determine the N concentration in dilute $GaAs_{1-x}N_x$ films ($x_{XRD} \leqslant 0.055$).

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1. Introduction

$GaAs_{1-x}N_x$ and $In_yGa_{1-y}As_{1-x}N_x$ have attracted interests because of their unique electronic and optical characteristics. It is well known that the incorporation of a small amount of N in $GaAs_{1-x}N_x$ leads to a decrease of the bandgap energy due to the large bandgap bowing, and to an increase of the electron effective mass. These alloys have been studied because of their potential applications in long-wavelength optoelectronic devices [1,2] and high-efficiency multijunction (MJ) solar cells [3]. A great deal of work has been done on the epitaxial growth and characterization of the III–V-N-type alloys in order to control the bandgap and electrical properties by controlling the alloy composition. While the bandgap and lattice constants as a function of the alloy composition for $GaAs_{1-x}N_x$ have been studied [1–4], further work is obviously necessary to investigate the micro-(nano-) structural and optical properties of $GaAs_{1-x}N_x$ as a function of N concentration and determine the alloying effect of GaN on GaAs.

Raman spectroscopy is a powerful technique to obtain information on crystal structure through measuring the vibrations of the crystal lattice. Raman spectra provide a sensitive tool for studying the impurity incorporation in such structure and the structural defects [5]. It is known that N-related local vibrational mode (LVM) absorption is directly proportional to the N concentration in $GaAs_{1-x}N_x$. Seong et al. compared the N concentrations in $GaAs_{1-x}N_x$ determined by Raman spectroscopy technique (x_{Raman}) with those determined by X-ray diffraction (XRD) (x_{XRD}) [4]. They demonstrated that x_{Raman} exhibits a linear dependence on x_{XRD} for $x_{XRD} < 0.03$ and some deviation from the linear dependence for $x_{XRD} > 0.03$. In this paper, we first review the results of Raman

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spectroscopic studies on the local bonding of N in $GaAs_{1-x}N_x$ with N concentrations up to $x_{XRD} = 0.055$. Second, we describe the N concentration dependence of the LVM in $GaAs_{1-x}N_x$ films.

2. Experiments

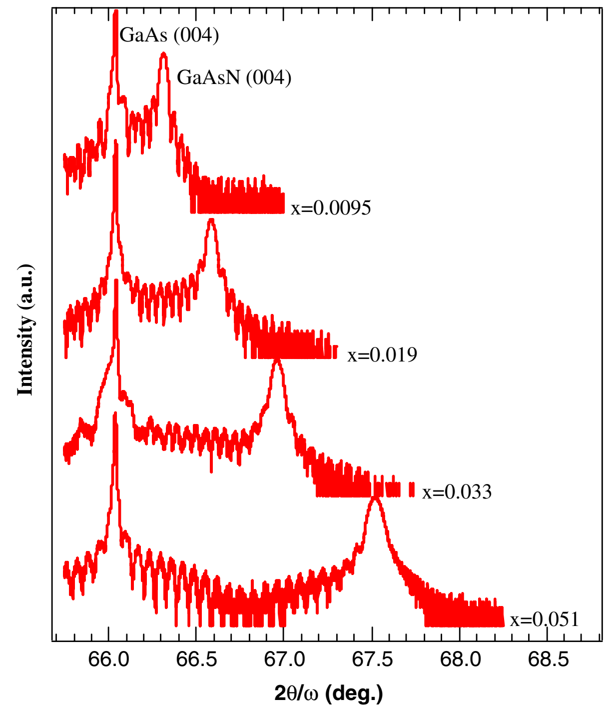
2.1. Procedure

$GaAs_{1-x}N_x$ films were grown by low-pressure (60 Torr) MOVPE using trimethylgallium (TMG), dimethylhydrazine (DMHy) and tertiarybutylarsine (TBAs) as the source materials of Ga, N and As, respectively. Ultra-high purity H_2 was used as a carrier gas at flow rate of 1.5 slm. All GaAsN films were nominally un-doped and grown on semi-insulating (SI) GaAs (0 0 1) substrates at temperature ranging from 475 to 600 °C. An un-doped GaAs buffer layer of ~300 nm was first grown at 650 °C. The substrate temperature was then reduced to 450–600 °C for the growth of GaAsN layer (200–300 nm). The [TBAs]/[TMGa] ratio was optimized to be 15. The flow rates of TMG and TBAs were fixed at 8.64 and 129.70 μmol/min, respectively. The flow rate of DMHy was varied in the range of 0–7000 μmol/min. N content in the GaAsN films was determined from a symmetrical (0 0 4) and an asymmetrical (1 1 5) high-resolution X-ray diffraction (HRXRD), assuming a linear dependence of lattice constant on the nitrogen concentration [6].

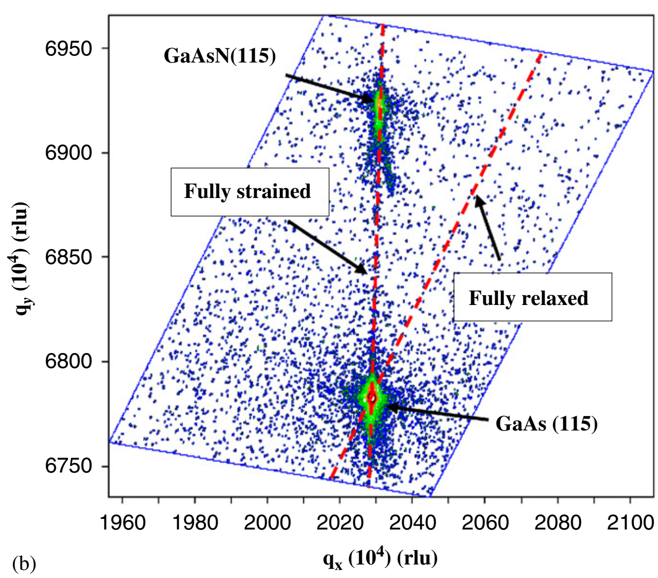
Micro-Raman scattering measurements were performed at room temperature (RT) using 514.5-nm-line Ar^+ laser as an excitation light source in backscattering geometry. The excitation laser beam was focused by a microscope lens system yielding a spot size ~2–4 μm in diameter.

3. Results and discussion

All $GaAs_{1-x}N_x$ films ($0 \leqslant x_{XRD} \leqslant 0.055$) in this study were examined by HRXRD in order to determine the N concentration and verify the structural quality. Fig. 1a shows a symmetric (0 0 4) HRXRD scans from a set of the $GaAs_{1-x}N_x$ films with different N contents. As shown in the figure, compared with GaAs, the peak shift to the higher diffraction angles was clearly observed with increasing N concentration, indicating the decrease of a lattice constant ($a_\perp$) normal to the (0 0 1) surface with the N incorporation. The narrow diffraction peaks and the clear Pendellösung fringes indicate that GaAsN films with high crystal quality were coherently grown on the GaAs substrate. An example of the reciprocal lattice mapping of an asymmetrical (1 1 5) reflection is shown in Fig. 1b for the $GaAs_{1-x}N_x$ film with N content of $x_{XRD} = 0.051$. This confirms that such high N-containing GaAsN layer is coherently strained and is of good epitaxial quality. These results are the evidence of high crystal quality films with higher N contents up to ~5%, obtained by the enhanced incorporation of N at lower growth temperatures (~500 °C) in spite of a large miscibility gap [7].



(a)



(b)

Fig. 1. (a) High-resolution (0 0 4) X-ray diffraction profiles of $GaAs_{1-x}N_x$ layers grown on the GaAs (0 0 1) substrates. Reciprocal lattice maps of the (1 1 5) reflection of the $GaAs_{1-x}N_x$ film with $x_{XRD} = 0.0051$ is shown in (b) as an example.

Fig. 2 shows typical Raman spectra recorded at room temperature (RT) for the $GaAs_{1-x}N_x$ film with N content of $x_{XRD} = 0.041$. Raman spectrum shows the transverse optical (TO) and longitudinal (LO) phonon modes of GaAs, along with a single N-related LVM. It is known that the Raman spectra taken from the GaAsN layer shows the GaN-like LO phonon mode, namely N-related LVM, at 470 cm^{-1} [8–10].

A sequence of room temperature Raman spectra taken from the $GaAs_{1-x}N_x$ films with N contents up to $x_{XRD} = 0.051$ is displayed in Fig. 3. The Raman intensity