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Optical properties of InN films grown by molecular beam epitaxy at different conditions

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Abstract

InN films were grown by N_2 plasma-assisted molecular beam epitaxy on Al_2O_3 substrates with GaN buffer layers at different substrate temperatures from 200 to 500 °C. It was found that the crystal quality of InN films was improved with growth temperature. The optical absorption edge of InN films decreased from 1.8 to 1.1 eV with increasing substrate temperature from 200 to 500 °C. Photoluminescence measurement on InN films grown at 500 °C exhibited the band-edge emission at around 1.0–1.1 eV.

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1. Introduction

Since InN has the smallest effective mass and the highest electron drift velocity among nitride semiconductors, this material is a most promising material for high-speed and high-frequency electronic devices [1]. However, InN remains most mysterious compound mainly due to difficulty in growing high-quality crystals. Currently, InN has been attracting strong attention, since new band-gap energies of 0.7–1.0 eV [2–4] have been recently reported well below the previously reported values of 1.8–2.0 eV [5]. Recently, Shubina et al. [6] claimed that a value of E_g lower than 1 eV was due to Mie resonance, while the infrared emission at around 0.7 eV from InN was associated with surface/gap states. On the other hand, F. Bechstedt et al. [7] commented that the band gap of InN was below 1 eV. Quite recently, Guo et al. [8] observed strong photoluminescence (PL) at 1.87 eV in InN films together with a clear absorption edge at 1.97 eV, but the quality of their films were not good. All of those studies suggest that the band-gap value of InN films is strongly dependent on the growth condition. However, little has been reported on how the growth conditions influence the crystal quality of InN films.

In this paper, we will present a study on the impact of growth conditions on optical properties of InN layers grown on Al_2O_3 substrates by radio frequency plasma-assisted molecular beam epitaxy (MBE). It is found that, as substrate temperature is raised from 200 to 500 °C, the crystal quality of InN films is improved, while the optical absorption edge decreases from 1.8 to 1.1 eV. PL measurement on InN films grown at 500 °C shows the band-edge emission at around 1.0–1.1 eV.

2. Experimental details

InN films were grown by N_2 plasma-assisted molecular beam epitaxy (MBE) on Al_2O_3 substrates. The Al_2O_3 substrates were ultrasonically cleaned in acetone and methanol each for 10 min prior to the oxide removal by a mixture of H_2SO_4/H_3PO4 (3:1) at 150 °C for 10 min. After loading into the MBE growth chamber, the sample was outgassed at 800 °C for 20 min, followed by exposure to nitrogen plasma at 700 °C for 50 min.

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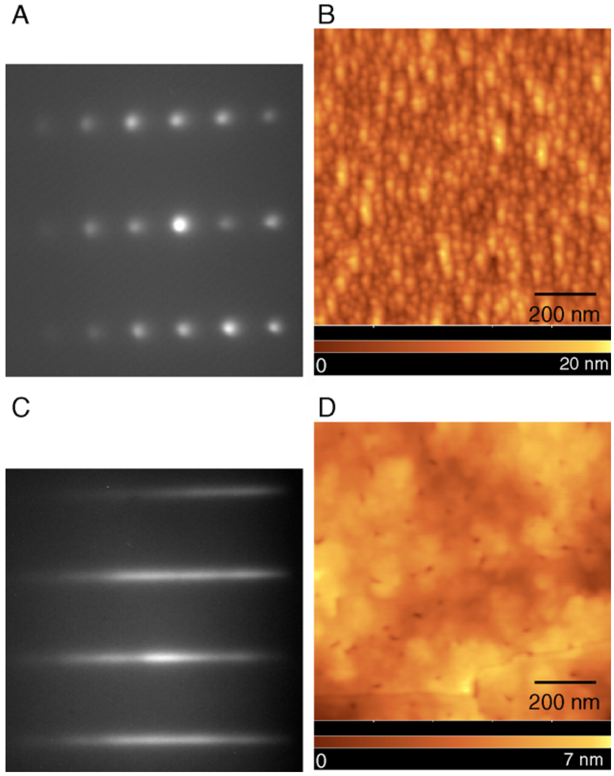

Fig. 1. (A, C) RHEED patterns and (B, D) AFM images (1 μm × 1 μm) of InN films grown on a c-Al_2O_3 substrate at 200 °C and 500 °C.

The nitrogen plasma source was operated at 250 W with a nitrogen flux resulting in the system pressures of 6.6×10^{-3} Pa (for nitridation of Al_2O_3 and GaN buffer layer growth) and 2.7×10^{-3} Pa (for InN growth). A low-temperature GaN buffer layer was grown at 500 °C for 3 min followed by annealing up to 900 °C for 15 min. The second GaN buffer layer was grown at 700 °C, followed by the growth of an InN film for 120–240 min at substrate temperatures of T_s from 200 °C to 500 °C. The InN samples were grown with Ga and In cell temperature at 900 °C and 835 °C, respectively. The growth rate of the InN films is about 2 nm/min.

The microstructure and crystal quality of InN films were characterized by in-situ reflection high-energy electron diffraction (RHEED) and high-resolution double crystal X-ray diffractometer (XRD) (Philips X'Pert-MPD). A monochromator containing Ge four crystals is used to obtain a narrow-band Cu-Kα_1 X-ray beam at the wavelength of 1.541 Å. The surface morphology was characterized by atomic force microscopy (AFM) (Digital Instruments NanoScope III) operating in contact mode with an Si_3N_4 tip. The Raman scattering experiments were performed on the Jobin-Yvon LabRam-INFINITY micro-Raman system at room temperature. A Kr^+ ion laser emitting at 514.5 nm together with a liquid nitrogen cooled Ge photodetector was used for the PL measurements from 10 K to 300 K. The optical absorption spectra were measured at 300 K using a JASCO's V-570 ultraviolet (UV)–visible (VIS)–near infrared (NIR) grating spectrophotometer. The resolution of the spectrophotometer is 0.1 nm and 0.5 nm at the range of UV/VIS and NIR, respectively.

3. Results and discussion

XRD and RHEED were used to characterize the crystalline structure of the grown films. Wurtzite-type InN [0002] and [0004] peaks were clearly observed, indicating that a highly *c*-axis oriented InN film was formed. No peak from In was observed. This show the InN grew without forming detectable In precipitation. All growth processes were monitored by RHEED. Usually, a spotty RHEED pattern of InN films will be observed during the growth of the films with lower T_s (200–300 °C), indicating 3D growth of the films. The RHEED pattern and AFM image of InN film grown at 200 °C were showed in Fig. 1(A) and (B). While streaky RHEED pattern will appear soon after the growth of InN layer with higher T_s (T_s = 400–550 °C) on GaN buffer layer and will keep until the end of growth. Fig. 1(C) and (D) shows the RHEED patterns and AFM image of InN film grown at 500 °C. The root-mean-square values of the surface of the InN layer is only about 0.5 nm. The streaky RHEED patterns and smooth surface of the film indicates two-dimensional growth of InN films. From RHEED and AFM measurement of the all InN films, no visible In precipitation was observed.

Fig. 2 shows Raman spectra of InN films with different growth temperature T_c observed by backscattering from the grown surface using the Ar^+ laser at 514.5 nm. Except for weak phonon signals from the GaN buffer layer and Al_2O_3 substrate, the spectral features of InN with T_s = 500 °C resemble very well with those of the InN films reported [3,9] in the sense that phonon modes A_1 (LO) at 587 cm^{-1} and E_2 high-frequency mode at 488 cm^{-1}. A LO-phonon-plasmon-coupled mode

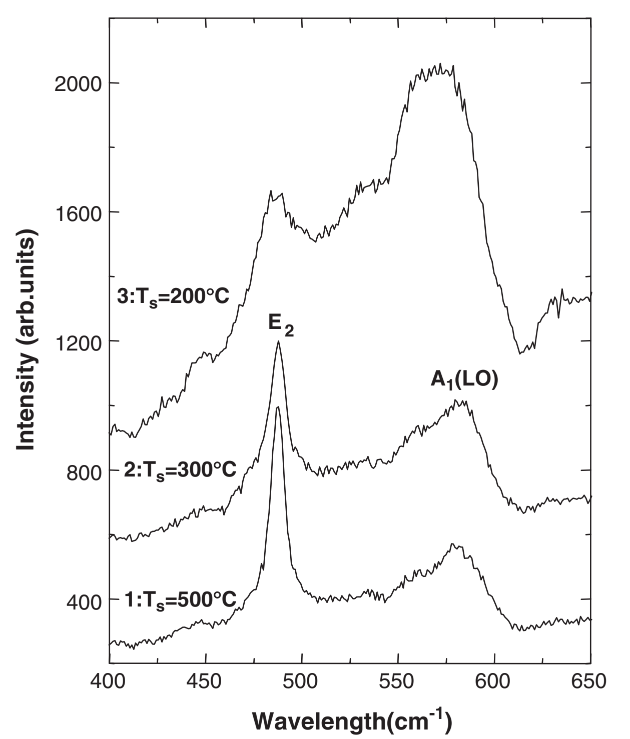

Fig. 2. Raman spectra of InN films with substrate temperatures T_s = 200, 300 and 500 °C.