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High nitrogen composition–induced low interfacial roughness of $GaAs_{0.978}N_{0.022}$/GaAs multiple quantum wells grown through solid-source molecular beam epitaxy

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ABSTRACT

$GaAs_{1-x}N_x$/GaAs multiple quantum wells (MQWs) were grown on GaAs(001) substrates through solid-source molecular beam epitaxy under various nitrogen background pressures (NBPs), and the crystal quality at the interface of $GaAs_{1-x}N_x$ and GaAs was investigated. X-ray diffraction and electron microscopy confirmed the low interface roughness of MQWs grown at a NBP of 5×10^{-6} Torr. Surface morphology measurements revealed a smooth surface without whisker-like defect structures. The fabricated MQWs exhibited high photoluminescence intensity because of the reduction in surface recombination with high nitrogen incorporation. Raman spectroscopy confirmed the presence of N-like local vibrational mode, and this was attributed to the presence of phase separation in GaAsN alloys. Rapid thermal annealing improved photoluminescence intensity by 100-fold and substantially reduced full width at half maximum because of MQW homogenization. These results evidence the favorable crystal interface of $GaAs_{0.978}N_{0.022}$ alloys. Hence, $GaAs_{0.978}N_{0.022}$/GaAs MQWs grown under high pressure might be useful in fabricating optoelectronic devices.

1. Introduction

Unlike conventional compound semiconductors such as GaInAs, AlGaAs, and GaInP, whose band gap energy can be approximated as a linear weighted average of the band gaps of the parental binary semiconductors, nitrogen-containing III–V compound semiconductor alloys exhibit large band gap bowing on incorporation of small amounts of nitrogen and have been extensively investigated in recent years [1,2]. InGaAsN containing 3% N and 9% In are ideal candidates for photovoltaic applications in multijunction solar cells owing to their bandgap of approximately 1 eV. These alloys can be lattice matched to GaAs or Ge substrates [3]. The efficiency of multijunction solar cells containing InGaAsN sub-cells is higher than 43%. The large difference in the atomic sizes of As and N induces a large miscibility gap in GaAsN, which in turn causes the phase separation of GaN, GaAsN, and GaAs at near-equilibrium [4,5]. Several growth techniques, such as chemical beam epitaxy [6], metalorganic molecular beam epitaxy (MBE) [7,8], metal–organic vapor-phase epitaxy [9], plasma-assisted MBE [10], and radio-frequency solid-source MBE (SS-MBE) [11], have been used to fabricate alloys with favorable characteristics without phase separation. These methods facilitate chemical reactions at the substrate surface, far from the equilibrium.

Coherently strained GaInAsN/GaAs MQWs have been widely used to fabricate long-wavelength vertical-cavity surface-emitting lasers (VCSELs). For example, Ellmers et al. fabricated photo-pumped 1.3-μm-wavelength VCSELs [12] and Kondow et al. fabricated 1.3-μm-wavelength QW edge-emitting lasers [13]. However, the surface segregation of In atoms and diffusion cause interface broadening in GaInAsN/GaAs MQWs [14]. Flat and abrupt interfaces are essential for fabrication of high-performance devices. Because of >20% lattice mismatch of GaAs and GaN, nitrogen incorporation generates tensile strain in GaAsN, which results in structure relaxation through the formation of misfit dislocations at the interface when the epilayer exceeds the critical thickness [15,16]. In addition, the strained layer releases strain-

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forming three-dimensional islands, which degrade the structural and optical properties of the heterostructures [17]. Tensile strain in MQWs/superlattices and thermal resistance of the interfaces are highly sensitive to thermal conductivity [18–20]. Interface defects probe phonon scattering [21], and the structural disorder in low dimensional material affects the vibrational energy transport [22]. However, the surface may start roughening even when the critical thickness is not exceeded [23]. Given these drawbacks, GaAsN/GaAs MQWs have not been extensively studied. Mussler et al. investigated the effect of well thickness on the roughening of GaAsN/GaAs MQWs with high nitrogen content [17]; however, the effect of NBP on MQWs is yet to be investigated. Random and inhomogeneous distribution of nitrogen atoms drastically degrades the optical characteristics of these alloys. Usually, post-growth thermal annealing is performed to improve the luminescence properties, which blue-shifts the photoluminescence (PL) peak energy because of alloy homogeneity [24].

Therefore, for realizing favorable crystal quality and an abrupt heretointerface, we investigated the effects of NBP on the growth of GaAsN/GaAs MQWs through SS-MBE. Further, we study the vibrational properties of the heterostructures to examine phase separation in GaAsN alloys. The results are expected to enhance our understanding of the interface crystal quality of $GaAs_{1-x}N_x$/GaAs MQWs and their application in optoelectronic devices.

2. Experimental

Five-period $GaAs_{1-x}N_x$/GaAs MQWs were grown using an SS-MBE system (DCA-450) equipped with an RF plasma source (SVT Associates) for incorporating atomic nitrogen in GaAsN QWs; the system was operated at 13.56 MHz for transforming molecular nitrogen into atomic nitrogen. The purity order for Ga and As_4 was 6N and that for N_2 was 6N5. Samples were grown on semi-insulating GaAs(001) substrates. Native oxide desorption was performed under As_2 flux at 600 °C, and a clear streaky pattern was obtained. At a beam equivalent pressure of approximately 4×10^{-8} Torr, a 250-nm GaAs buffer layer was grown at 580 °C on the GaAs substrates; a clear 2 × 4 surface reconstruction was observed on the RHEED screen. At a ramp rate of 30 °C/min, the substrate temperature was then reduced to 490 °C for growing five-period GaAsN/GaAs MQWs. Immediately after growing the buffer layer, plasma was ignited and stabilized at a forward power of 450 W with the plasma shutter closed. Subsequently, two five-period $GaAs_{1-x}N_x$/GaAs MQWs were grown at NBPs of 4×10^{-6} and 5×10^{-6} Torr with the plasma shutter open during GaAsN QW growth. The shutter was closed immediately after growing the final nitride layer. The growth rate of both GaAs and GaAsN layers was nearly 1 μm/h. The projected thicknesses for the GaAsN QW and GaAs barrier layer were 5.5 nm and 50 nm, respectively. A schematic of the dilute nitride heterostructure is illustrated in Fig. 1.

The structural properties of the MQWs, such as interface crystal quality, interface roughness, composition, and strain were investigated through high-resolution X-ray diffraction (HR-XRD, Rigaku) by using a double-axis diffractometer. Surface morphology was studied through field-emission gun-scanning electron microscopy (FEG-SEM, JOEL). The optical properties of the MQWs were investigated through PL spectroscopy by using a 532-nm diode pump solid-state laser with a 25-mW excitation source. The PL measurements were conducted in a closed-cycle helium cryostat at 19 K and an InGaAs signal detector. The interface crystal quality of the MQWs was verified through transmission electron microscopy (TEM; JEOL 300). The vibrational properties of the MQWs were examined through room-temperature Raman spectroscopy (Horiba HR 800). Finally, to improve optical

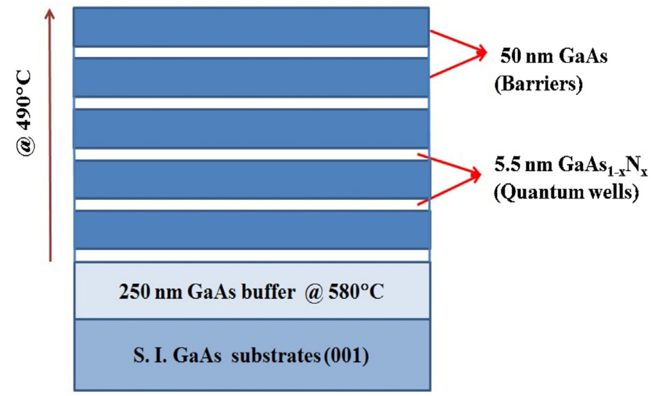

Fig. 1. Schematic of the five-period GaAs1−xNx/GaAs MWQs grown at NBPs of (a) 4×10^{-6} and (b) 5×10^{-6} Torr.

properties, the samples were subjected to rapid thermal annealing (RTA; Annealsys As One) for 30 s at 750 °C in nitrogen ambient.

3. Results and discussions

3.1. Structural properties

The hetero-interface quality of the five-period $GaAs_{1-x}N_x$/GaAs MQWs grown at two NBPs were analyzed by performing HR-XRD ω–2θ scan around the symmetric (004) reflection of GaAs. The N compositions in the MQWs grown at NBPs of 4×10^{-6} and 5×10^{-6} Torr were 1.8% and 2.2%, respectively. The highest intensity peak was observed at 66° (Fig. 2(a)) and was attributed to the GaAs layer. Satellite (SL) peaks were designated as −1, −2, −3, −4, 1, and 2; the zero-order (0) SL peak was found on the shoulder of GaAs (004) at a low angle. The zero-order SL peak for the two samples was observed at different 2θ values because the generated tensile strain is dependent on the level of N incorporation in the QWs. Fig. 2(b) presents the full width at half maximum (FWHM) of the SL peaks of the MQWs as a function of the order of the SL peak. The FWHM of the higher-order SL peaks broadened as the N concentration in the MQWs increased. SL peak broadening due to intermixing and/or irregularity in QW thickness was attributed to variations in alloy composition [25]. The interface quality was investigated by employing approaches similar to those used for InGaN/GaN [26,27] and InGaAs/GaAsP MQWs [28]. Interface roughness strongly affects SL peak broadening. If the interface roughness appears due to periodical thickness-induced fluctuations, described by a Gaussian distribution with standard deviation σ, the FWHM of nth order SL peak can be expressed using the following expression [28]:

$$\beta_n = \beta_0 + (\ln 2)^{1/2}\, n\, \Delta\theta_M.\, \sigma/\Lambda \quad (1)$$

where Λ is the periodicity of the system, n is order of the SL peaks, $\Delta\theta_M$ is the difference in angle between the two adjacent SL peaks, and β_n and β_0 are the FWHM of the n- and zero-order SL peak, respectively. $\Delta\theta_M$ of the samples differed slightly, possibly because of the minor variations in well thicknesses due to random nitrogen incorporation. As the well thickness varies slightly, so also the thickness of period Λ (well + barrier) does, leading to a variation in $\Delta\theta_M$ [29]. The FWHM of the SL peaks and the SL order peak exhibited a nearly linear relationship. From the slope coefficient $[(\ln 2)^{1/2}\, \Delta\theta_M\, \sigma/\Lambda]$ of the linear fit, the interface roughness (σ/Λ) was calculated and it was 3.23% and 2.02% for the $GaAs_{0.983}N_{0.018}$/GaAs and $GaAs_{0.979}N_{0.022}$/GaAs MQWs, respectively. The former exhibited hazy interfaces indicating a relatively poor-quality interface, whereas sharp and abrupt interfaces were observed in