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Interdiffusion in narrow InGaAsN/GaAs quantum wells

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Interdiffusion in $\text{In}_{0.32}\text{Ga}_{0.68}\text{As}_{0.984}\text{N}_{0.016}/\text{GaAs}$ multiple quantum wells with well widths of 2 and 4 nm, respectively, was investigated both experimentally and theoretically. Maximum blueshifts of 206 and 264 meV in the photoluminescence spectra were observed. Secondary ion mass spectrometry showed that both In–Ga and N–As interdiffusions played key roles for the large blueshifts. The significant In–Ga interdiffusion occurred at 650 °C while the N diffusion occurred at a temperature above 700 °C. The theoretical results are in good agreement with the experimental observations. © 2007 American Institute of Physics. [DOI: 10.1063/1.2736943]

I. INTRODUCTION

In recent years InGaAsN alloy has been attracting great attention owing to the unique nitrogen-driving physical and optical properties and the possible applications for optoelectronic devices, such as semiconductor lasers for optical communications,^{1–3} solar cells with very high internal quantum efficiencies,⁴ and infrared photodetection.^{5–7} It has been found that the introduction of a small amount of nitrogen (N) into (In,Ga)As reduces the band gap greatly and increases the electron effective mass significantly. They are attributed to the band anticrossing (BAC) effect resulting from the interaction of the conduction band and the high-lying N resonant state although the exact physical mechanism behind remains a subject of ongoing debates.^{8–11}

A postgrowth rapid thermal annealing (RTA) is often required for InGaAsN based quantum wells (QWs) to ameliorate the poor crystal quality due to the incorporation of nitrogen and improve the material properties. It has been commonly observed that InGaAsN/GaAs QWs show some blueshifts of emission wavelength after RTA processing.^{12–21} However, the detailed fundamental behind the blueshift phenomenon was under debate, and several possible mechanisms have been proposed. For example, Sun *et al.* and Kageyama *et al.* suggested In–Ga interdiffusion,^{12–14} Albrecht *et al.* suggested N diffusion out of the QWs,¹⁵ Pan *et al.*, Li *et al.*, and Spruytte *et al.* observed both In and N diffusions,^{16–18} and Kurtz *et al.* and Duboz *et al.* supported the change in N-bonding configuration from a Ga-rich to an In-rich environment.^{19,20} In addition, Oye *et al.* have investigated In intra- and interdiffusion mechanisms in InGaAs/GaAsN QWs with RTA processing and found that a lot of N atoms diffused from the GaAs barriers into the InGaAs wells.²¹

In this article, we report the observation of very large blueshifts resulted from great In–Ga and N–As interdiffusions in the $\text{In}_{0.32}\text{Ga}_{0.68}\text{As}_{0.984}\text{N}_{0.016}/\text{GaAs}$ multiple quantum

wells (MQWs) with narrow well width. A theoretical study based on the ten-band $\mathbf{k}\cdot\mathbf{p}$ model is also carried out to analyze the subband dispersions in the InGaAsN QWs with the diffusion-induced complex potential profiles.

II. EXPERIMENT

Two InGaAsN/GaAs samples were grown by a RIBER-32P solid source molecular beam epitaxy (SSMBE) on double-side polished (100) semi-insulating GaAs substrates under the same growth conditions. Nitrogen was provided by a radio-frequency plasma source, arsenic was provided by a valved cracker cell, and the rest elements were provided by thermal effusion cells. Sample 1 (S1) consists of eight periods of 2 nm $\text{In}_{0.32}\text{Ga}_{0.68}\text{As}_{0.994}\text{N}_{0.016}$ wells and 35 nm GaAs barriers. Sample 2 (S2) is identical to S1 except the 4 nm well width. A *n*-type GaAs buffer layer of 0.4 μm was grown first, followed by eight QWs and then a 0.2 μm GaAs cap layer. The growth temperatures were 440 °C for the QWs and 590 °C for the buffer and cap layers. Both the shutter of the ignited N plasma source and the N valve were closed during the growth of buffer, cap, and barrier layers to avoid N incorporation into the GaAs layers. Clear streaky reflection high-energy electron diffraction (RHEED) patterns were observed for both samples.

High-resolution x-ray diffraction (HRXRD) was employed to characterize the two InGaAsN/GaAs MQW structures. Figure 1 shows the (004) GaAs Bragg reflection rocking curves in the $\omega/2\theta$ geometry measured by using a Philips X'Pert materials research diffractometer. The solid curve is the experimental result, and the dash curve is the simulation based on the dynamical x-ray theory. The excellent agreement between them confirms the structural and composition parameters. The peak with the greatest intensity is from the GaAs substrate, and the satellite peaks are from the MQWs. It can be seen that well-defined periodic satellite peaks up to the 10th order for S1 and 14th order for S2 can be identified, indicating good layer periodicity. The sharpness of the peaks represents the abruptness of the interfaces of the strained MQWs. The angular spacing between the satellite peaks is

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FIG. 1. XRD (004) rocking curves of (a) S1 and (b) S2 together with the simulation results. The simulation curve was shifted vertically for clarity.

directly determined by the period of the MQWs. P_{GaAs} and $P0_{\text{satel}}$ show the positions of GaAs substrate (004) diffraction and the zero-order satellite peak, respectively. $P0_{\text{satel}}$ deviates from P_{GaAs} by about -1.33° , indicating a large compressive strain between the $\text{In}_{0.32}\text{Ga}_{0.68}\text{As}_{0.994}\text{N}_{0.016}$ well and the GaAs barrier.

The samples were cleaved into $3 \times 3 \text{ mm}^2$ squares and then coated with 120 nm thick SiO_2 cap layers which were deposited by plasma-enhanced chemical-vapor deposition (PECVD) system to prevent surface damage during annealing. RTA was performed in an ETSTAR 100 system for 60 s at temperatures of 650, 700, and 750 $^\circ\text{C}$, respectively. During the thermal processing, the samples were kept in flowing nitrogen ambient for protection. For the photoluminescence (PL) measurement, a 532 nm laser was used as the exciting source and an InGaAs photodetector was used to detect the PL signal.

III. RESULTS AND DISCUSSION

Figure 2 shows measured PL spectra under different annealing conditions. For S1, the as-grown sample exhibits an emission peak at 1235.9 meV. It shifts to 1337.6, 1419.6, and 1441.6 meV after RTA for 60 s at 650, 700, and 750 $^\circ\text{C}$, respectively. For S2, the as-grown sample shows an emission peak at 1078.9 meV. It shifts to 1173.1, 1316.5, and 1342.6 meV after annealing for 60 s at the three temperatures, respectively. It can be seen that the blueshifts increase rapidly with annealing temperature and then tend to saturate when the temperature is above 700 $^\circ\text{C}$. The maximum blueshifts of 205.7 and 263.7 meV for S1 and S2, respectively,

FIG. 2. PL spectra measured at 77 K for the two InGaAsN/GaAs MQWs, (a) S1 and (b) S2, with different thermal annealing conditions: (1) as grown, (2) 650 $^\circ\text{C}$, (3) 700 $^\circ\text{C}$, and (4) 750 $^\circ\text{C}$. The annealing time is 60 s.

were observed with RTA processing at 750 $^\circ\text{C}$. To our knowledge, they are the largest blueshifts reported so far for InGaAsN based QWs. The RTA processing has resulted in some improvement on the material quality. For instance, the full width at half maximum (FWHM) of the PL spectrum in S1 is decreased from 62.8 to 40.0, 32.9, and 28.7 meV, after being annealed at 650, 700, and 750 $^\circ\text{C}$, respectively. This FWHM narrowing is also observed in S2 although it is not as significant as in S1. In addition, S1 and S2 both show larger PL peak intensity after being annealed at 750 $^\circ\text{C}$.

To examine the cause of the great blueshifts in PL spectra, an analysis of composition profiles was done using secondary ion mass spectrometry (SIMS). Figures 3 and 4 show the results of In, Ga, and N along the growth direction in sample S2 with different RTA conditions. The nonsquare shape of the composition profile in the SIMS spectrum for the as-grown MQW is mainly due to the limited depth resolution and the ion beam mixing effect. Although the composition profiles obtained with SIMS are relatively rough, they clearly revealed that not only In–Ga interdiffusion but also N outdiffusion from the narrow wells occurred. The average In composition in the InGaAsN wells is about 32% for the as-grown sample, while the peak composition reduces to about 23%, 21%, and 19% for samples annealed at 650, 700, and 750 $^\circ\text{C}$, respectively. The diffusion of N is more severe. Its average composition in the InGaAsN wells is about 1.6% in the as-grown sample. The peak composition decreases quickly to about 1.1%, 0.62%, and 0.61% for the samples annealed at 650, 700, and 750 $^\circ\text{C}$, respectively. We can also observe that the variations of In and N diffusions with in-

FIG. 3. Composition profiles of Ga and In measured by SIMS for S2 under different RTA conditions: (a) as grown, (b) 650 °C, (c) 700 °C, and (d) 750 °C.

creasing RTA temperature show different trends. The In–Ga interdiffusion occurs significantly at 650 °C, but becomes slow when the temperature increases further to 700 and 750 °C. However, the diffusion of N is strong at about 700 °C and slows down at 750 °C. It indicates that the N diffusion tends to dominate at higher annealing temperatures.

The observed diffusion behaviors shown in Figs. 3 and 4 are consistent with the blueshifts in the PL shown in Fig. 2 for different RTA temperatures. The blueshift in the annealed sample at 650 °C should result from moderate In and N diffusions. The large increase of blueshift in the annealed sample at 700 °C is mainly due to the severe N diffusion. The slight increase of blueshift with the RTA temperature at 750 °C is a result of the remnant In and N diffusions.

The large In–Ga interdiffusion upon RTA can be explained by the impurity-free vacancy disordering (IFVD).^{22,23} The SiO₂ dielectric cap layer is known to induce outdiffusion of Ga atoms during annealing, which generates

FIG. 4. Composition profiles of N measured by SIMS for S2 with different RTA conditions: (a) as grown, (b) 650 °C, (c) 700 °C, and (d) 750 °C.

FIG. 5. Calculated dependence of In composition profile on diffusion length L_d in the $\text{In}_{0.32}\text{Ga}_{0.68}\text{As}_{0.984}\text{N}_{0.016}/\text{GaAs}$ QWs with well widths of (a) 2 nm and (b) 4 nm, respectively.

group III vacancies and results in large interdiffusion of group III atoms in the underlying MQW region. The significant reduction of N at about 700 °C is mainly due to the outdiffusion of nitrogen as it becomes volatile at high annealing temperature.

The narrow well widths in our InGaAsN/GaAs MQWs enhance the RTA-induced blueshifts. To account for this, we employ a classic expression to model the composition profile after interdiffusion in the QW as follows:²⁴

$$x'(z) = \frac{x}{2} \left[\operatorname{erf} \left(\frac{L_w + 2z}{4L_d} \right) + \operatorname{erf} \left(\frac{L_w - 2z}{4L_d} \right) \right], \quad (1)$$

where x is the as-grown composition, z is the distance in the growth direction, L_w is the well width before interdiffusion, and L_d is the diffusion length. Figures 5(a) and 5(b) show the calculated In composition profiles with different diffusion lengths for S1 and S2, respectively. At $L_d = 1$ nm, about 50% and 31% of the In content diffuse out from the original 2 and 4 nm wide well regions, respectively. At $L_d = 2$ nm, the diffusion proportions become about 73% and 52% for the two QWs, respectively. It indicates that the increase of the QW band gap or the PL energy in a narrower well should be larger than that in a wider well upon the same RTA processing. The blueshifted PL peak at 1441.6 meV for S1 with RTA at 750 °C is larger than the 1342.6 meV for S2, confirming the theoretical predication.

IV. THEORETICAL STUDY

It is favorable to do a detailed theoretical analysis about the dependence of QW emission peak on the composition diffusion to account for the experimental results. Two important facts should be considered in the analysis. One is the complex QW potential profile resulted from interdiffusion, and the other is the existence of high-lying N resonant states. Some theoretical work on the interdiffusion in InGaAsN/GaAs QWs has been reported.^{24,25} Chan *et al.* have investigated the group III interdiffusion using a one-dimensional Schrödinger-like equation to calculate the electron and hole subband energies.²⁴ Dang *et al.* have also done a theoretical study on the In–Ga interdiffusion with the eight-band $\mathbf{k} \cdot \mathbf{p}$ model.²⁵ In this section, we employed the ten-band $\mathbf{k} \cdot \mathbf{p}$ model²⁶ combined with the envelope function Fourier expansion²⁷ to calculate the eigenstates in the annealed

InGaAsN/GaAs QWs. The ten-band $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian which is based on the conventional eight-band $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian and the BAC theory can be described by

$$\mathbf{H} = \begin{bmatrix} \mathbf{H}_{p\bar{s}} & \mathbf{H}_{p\bar{s}-N}^* \\ (8 \times 8) & (8 \times 2) \\ \mathbf{H}_{p\bar{s}-N} & \mathbf{H}_N \\ (2 \times 8) & (2 \times 2) \end{bmatrix}, \quad (2)$$

where $\mathbf{H}_{p\bar{s}}$ is the eight-band Hamiltonian,²⁸ $\mathbf{H}_N$ is the N band Hamiltonian, and $\mathbf{H}_{p\bar{s}-N}$ consists of the coupling terms connecting the conduction and valence bands with the N band. $\mathbf{H}_N$ and $\mathbf{H}_{p\bar{s}-N}$ take the following forms:

$$\mathbf{H}_N = \begin{bmatrix} E_{N0} + \alpha_N k^2 & 0 \\ 0 & E_{N0} + \alpha_N k^2 \end{bmatrix}, \quad (3)$$

and

$$\mathbf{H}_{p\bar{s}-N} = \begin{bmatrix} V_{NC} & 0 & -\sqrt{3}T_{N+} & \sqrt{2}Q_N & -T_{N-} & 0 & -Q_N & -\sqrt{2}T_{N-} \\ 0 & V_{NC} & 0 & T_{N+} & \sqrt{2}Q_N & -\sqrt{3}T_{N-} & \sqrt{2}T_{N+} & -Q_N \end{bmatrix}, \quad (4)$$

where $V_{NC} = \beta\sqrt{x}$ with nitrogen composition x describes the coupling between the conduction band and the high-lying N band. The matrix elements $T_{N\pm}$ and Q_N should vary linearly with k , coupling the N band with the valence band.

There are two most important N-related band parameters in Eqs. (3) and (4), E_{N0} and β , which are the N resonant state position and its interaction intensity with the conduction band, respectively. They are given by²⁰

$$E_{N0} = 1.65 - 0.31x \text{ eV and } \beta = 2.4(1 - x) + 1.75x \text{ eV} \quad (5)$$

for a Ga-rich environment of N-bonding configuration (in the as-grown case), and

$$E_{N0} = 1.7125 - 0.31x \text{ eV and } \beta = 2.4(0.75 - x) + 1.75(x + 0.25) \text{ eV} \quad (6)$$

for an In-rich environment of N-bonding configuration (in the RTA case).

In the ten-band scheme, the total wave function can be written as

$$\Psi(r) = \sum_{j=1}^{10} F_j(r) u_j(r) = \sum_{j=1}^{10} \exp(ik_r \rho) \varphi_j(z) u_j(r), \quad (7)$$

where $F_j(r)$ is the envelope function, $u_j(r)$ is the periodic part of the Bloch basis function at the zone center, $k_r = (k_x, k_y)$ is the two-dimensional in-plane wave vector, and $\rho = (x, y)$ is the two-dimensional in-plane space vector. The Fourier expansion of the envelope function can be written as

$$F_j(r) = \exp[i(k_x x + k_y y)] \sum_m a_{j,m} \frac{1}{\sqrt{L}} \exp\left[i\left(k_z + m \frac{2\pi}{L}\right)z\right], \quad (8)$$

where m is an integer denoting the order of the related expansion term, L is the MQW structure period, and k_z is the wave vector in the growth direction. Substituting Eq. (8) into the coupled differential equations

$$\sum_{j'=1}^{10} [H_{jj'} + U(z) \delta_{jj'}] F_{j'}(r) = E F_j(r), \quad (9)$$

where $U(z)$ is the electronic potential function and $H_{jj'}$ is the ten-band $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian, then multiplying by $\phi_m^*(z)$ and integrating over L , one gets

$$\sum_{j',m'} H_{jj'}(m, m') a_{j',m'} = E a_{j,m}, \quad j, j' = 1, \dots, 10, \quad (10)$$

where the matrix elements $H_{jj'}(m', m)$ are given by

$$\begin{aligned} H_{jj'}(m, m') &= \int_{-L/2}^{L/2} \phi_m^*(z) [H_{jj'} + \delta_{jj'} U(z)] \phi_{m'}(z) dz \\ &= \langle \phi_m | H_{jj'} + \delta_{jj'} U(z) | \phi_{m'} \rangle. \end{aligned} \quad (11)$$

The electronic potential function $U(z)$ in Eq. (11) consists of two parts. One is the QW potential from the band discontinuity between the well and the barrier and the other is the strain-induced potential change. For the QWs before interdiffusion, both of them are in squarelike profiles, so the integration of $U(z)$ over L can be yielded analytically as

$$\langle \phi_m | U(z) | \phi_m \rangle = VL_B/L + \xi L_W/L,$$

$$\langle \phi_m | U(z) | \phi_{m'} \rangle = (-V + \xi) \frac{\sin(\pi L_W f)}{\pi f L} \quad (m \neq m'), \quad (12)$$

where V is the square QW potential and ξ is the strain-induced potential change. For the nonsquare QWs after interdiffusions, the integration cannot be done analytically and so the numerical method is required. The eigenenergies and eigenstates in the QWs can then be obtained by solving the giant algebraic set of coupled equations, i.e., Eq. (10).

To analyze the energy subband structures for S2 under different annealing conditions, the composition diffusion is modeled using Eq. (1). By fitting theoretical peak compositions upon interdiffusions to the measured SIMS results in Figs. 3 and 4, the In diffusion lengths are estimated to be 1.3,

FIG. 6. Calculated energy dispersions of conduction and valence subbands for S2 with estimated In and N diffusion lengths: (a) zero for as grown, (b) $L_d(\text{In})=1.3$ nm and $L_d(\text{N})=1.3$ nm for RTA at 650 °C, (c) $L_d(\text{In})=1.5$ nm and $L_d(\text{N})=2.8$ nm for RTA at 700 °C, and (d) $L_d(\text{In})=1.7$ nm and $L_d(\text{N})=2.9$ nm for RTA at 750 °C.

1.5, and 1.7 nm, and the N diffusion lengths are estimated to be 1.3, 2.8, and 2.9 nm, for the RTA processing at 650, 700, and 750 °C, respectively.

The basic III-V material parameters for the calculation were taken from Refs. 29 and 30. The N-related BAC parameters of E_{N0} and β were taken from Ref. 20. And the band offset parameters of $\text{In}_x\text{Ga}_{1-x}\text{As}_y\text{N}_{1-y}/\text{GaAs}$ were taken from Ref. 31. It is assumed that the high-lying N level only takes effect on the conduction band while its effect on the valence band can be neglected. Figure 6 shows the calculated energy dispersions of the conduction and valence subbands as a function of plane wave vector k_t along [100] and [110] directions for S2 with estimated diffusion lengths. Only the lowest three states are displayed in the figure. The separation between the ground state in the conduction band (E1) and the ground state in the valence band (HH1) at the zone center ($k_t=0$) is regarded as theoretical PL emission energy. The calculated E1-HH1 transition energies under different RTA conditions are plotted in Fig. 7 with the experimental PL values for comparison. It can be seen that the computed results are 1095 meV for the as-grown sample and 1186, 1304, and 1328 meV for the three samples annealed at 650, 700, and 750 °C, respectively. A good agreement between the theoretical and experimental PL emission peaks is achieved. The maximum discrepancy is less than 20 meV, which is likely due to the approximation of the composition profiles. The theoretical result confirms the influence of In and N diffusions on the PL emission energy in our samples. The In–Ga interdiffusion increases the well band gap directly, and the severe outdiffusion of N diminishes the BAC effect, resulting in an obvious blueshift in the PL.

V. SUMMARY

In conclusion, RTA-induced blueshifts of PL emission energy in two $\text{In}_{0.32}\text{Ga}_{0.68}\text{As}_{0.984}\text{N}_{0.016}/\text{GaAs}$ MQW struc-

FIG. 7. Comparison of the calculated E1-HH1 transition energies and the experimental PL emission peaks of S2 under different RTA conditions.

tures with well widths of 2 and 4 nm, respectively, have been studied both experimentally and theoretically. Maximum blueshifts of 206 and 264 meV have been observed in the two structures upon RTA at 750 °C for 60 s. It is found that the significant In–Ga interdiffusion occurred at 650 °C, while the severe N diffusion occurred at annealing temperature about 700 °C. The calculated E1-HH1 transition energies under estimated In and N diffusion lengths using the ten-band $\mathbf{k}\cdot\mathbf{p}$ model can account well for the experimental observations.

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