

Phonons and Valence-band Splitting in Strained GaAs_{1-x}N_x/GaAs Epilayers

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Abstract—The strain effects in strained GaAs_{1-x}N_x epilayers are characterized by Raman spectroscopy and photocurrent spectra at various nitrogen composition. In addition, the nitrogen composition and the strain were determined by using high-resolution X-ray diffraction (HR-XRD). The Raman spectra are observed to be dominated by the GaAs-like longitudinal optical (LO) phonon mode as the strongest peaks show up around 289~294 cm⁻¹. Moreover, the weak and broad peaks features in the range of 255~276 cm⁻¹ originate from the peaks of GaAs-like transverse optical (TO) phonon mode and disorder induced, GaN-like LO. And the Raman peak shifts toward lower wave number with increasing nitrogen compositions, which indicates the presence of tensile strain in the strained GaAs_{1-x}N_x epilayers. The valence-band splitting of GaAs_{1-x}N_x are obtained from photocurrent spectra. As the nitrogen concentration increases, the tensile strain in strained GaAs_{1-x}N_x epilayers increases while the valence-band splitting increases.

Index Terms—Phonon, band splitting, strain, GaAsN

I. INTRODUCTION

The GaAs_{1-x}N_x epilayer is of considerable importance today, because of its wide application in the optoelectronics device [1-3], and there have been many studies of its physical properties [4-6]. Such studies are

important because a proper understanding of the material characteristics underlies all applications. It is well known that the GaAs_{1-x}N_x epilayer is grown as a pseudomorphic layer on top of a GaAs buffer, the sizeable reduction of the lattice parameter of GaAs_{1-x}N_x due to nitrogen incorporation results in the development of a tensile strain [7-9]. The strain causes a phonon frequencies shift, valence-band splitting and an increase or decrease in the band gap under a compressive or tensile strain, respectively [10, 11].

Raman spectra are valuable tools for characterizing the lattice vibrational properties of bulk semiconductors and epilayer systems. Once the phonons in an alloy system have been characterized, Raman spectroscopy can be used, for example, to evaluate the alloy strain in a given situation from measurements of the optical phonon frequencies. Raman scattering has distinct advantages in many cases, as it may be universally applied to thin alloy epilayers such as quantum wells or to quantum wires and dots. There have been many Raman investigations of the GaAs_{1-x}N_x semiconductor alloy, principally in the form of strained epilayers grown on GaAs. Valence-band splitting in GaAs_{1-x}N_x are also reported [12, 13], but a theoretical analysis of these phenomena has not been carried out. Accordingly, detailed theoretical calculations of tensile strain effects in strained GaAs_{1-x}N_x epilayers are necessary and important. The fundamental band gap, valence-band splitting and spin-orbit splitting of GaAs_{1-x}N_x at various temperatures are measured by photoreflectance spectroscopy [14]. For photocurrent spectra, the low energy cut-off of the photocurrent spectra was determined by using the band gap or the absorption edge of the semiconductor [15]. A low energy cut-off was also observed in the photocurrent spectra characteristics

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because at low energy, the value of the absorption coefficient is very large in semiconductors and all the incident optical absorption is near the surface. The photocurrent spectra peaks are dominated by an intense band to band transition that is attributed to optical transition of the valence band electrons [15]. Therefore, by measuring the photocurrent spectra peaks we can distinguish between the exciting transitions involving light or heavy hole band in the $\text{GaAs}_{1-x}\text{N}_x$ epilayer. Thus, analysis of photocurrent spectra peaks shows that the light- and heavy-hole of valence-band in strained $\text{GaAs}_{1-x}\text{N}_x$ epilayer grown on GaAs substrates are split at the Γ point, with the valence-band of light- and heavy-hole, induced by tensile strain in $\text{GaAs}_{1-x}\text{N}_x$ epilayer. The photocurrent spectra are found to consist of two peaks at near band edge whose splitting increase with increasing nitrogen composition.

In this paper, we analyze the effect exerted by strain on the phonon and the valence-band splitting in strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers grown on GaAs (100) substrates, using Raman and photocurrent spectra measurements. In addition, the composition was determined by using high-resolution X-ray diffraction (HR-XRD) and Vegard's rule. All epilayers exhibit coherent tensile strain on the GaAs buffer layer. The Raman spectra are observed to be dominated by the GaAs-like longitudinal optical (LO) phonon mode as the strongest peaks show up around 289~294 cm^{-1} . Moreover, the weak and broad peaks features in the range of 255~276 cm^{-1} originate from the peaks of GaAs-like transverse optical (TO) phonon mode and disorder induced, GaN-like LO. The fundamental band gap and valence-band splitting of $\text{GaAs}_{1-x}\text{N}_x$ at various temperatures are obtained by photocurrent spectra. The photocurrent spectra show the valence-band splitting peaks, and the valence-band splitting increase with increasing nitrogen composition in the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayer.

II. EXPERIMENT

The strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers were grown by using metalorganic chemical vapor deposition (MOCVD) with triethylgallium (TEG), arsine (AsH_3), and dimethylhydrazine (DMHy) in a hydrogen carrier gas. The substrate temperature was 550 °C, the reactor pressure was 60 Torr, and the growth rate was 2.5 $\mu\text{m}/\text{h}$

for all strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers. First, a 0.1- μm -thick undoped GaAs buffer layer was grown on a (001) GaAs substrate, then, 0.8- μm -thick strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers were grown. When TEG flow was fixed at 40 $\mu\text{mol}/\text{min}$, the nitrogen composition increases with both increasing DMHy flow and decreasing growth temperature. By changing the flow of DMHy from 300 to 1500 $\mu\text{mol}/\text{min}$ at a growth temperature of 550 °C, the nitrogen composition was varied from $x = 0.29\%$ to $x = 0.61\%$. After strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers growth, a 0.02- μm thick GaAs cap layer was grown. The composition and the strain were determined by using high-resolution X-ray diffraction (HRXRD). The room-temperature Raman spectra of the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers were measured using a pumped solid-state laser unit of 532 nm. The spectra were taken at room temperature in a (001) back-scattering geometry for various nitrogen compositions. The band-gap was measured at temperatures ranging from 300 to 30 K by using photocurrent measurements. For the photocurrent measurements, the sample was cut into $3 \times 3 \text{ mm}^2$, and two coplanar electric contacts with a 1-mm spacing were formed with indium solder. The ohmic contact was confirmed using the I-V characteristics. A quartz-tungsten halogen lamp was used as a photo-excitation light source. The photocurrent spectrum was analyzed by a 500-mm grating monochromator. The photocurrent signal was picked up with a lock-in amplifier and then recorded using a computer. A bias of 0.5 V was supplied by a current source using a Keithley 236 source measurement unit for the conductivity measurements.

III. RESULTS AND DISCUSSION

The nitrogen composition in the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers was determined from X-ray diffraction and Vegard's rule. Fig. 1 shows the HRXRD rocking curves for a set of strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers with different nitrogen compositions in the range $0.29 < x < 0.61$. The well-defined diffraction peaks are regular with the growth of single-phase $\text{GaAs}_{1-x}\text{N}_x$ strained layers. With nitrogen composition increasing, the diffraction peak corresponding to $\text{GaAs}_{1-x}\text{N}_x$ strained layer shifts away from the GaAs substrate peak, which can be expected from the increasing lattice mismatch strain. $\text{GaAs}_{1-x}\text{N}_x$ strained layers are under tensile strain on GaAs

substrates. If tetragonal alteration is assumed, the lattice parameters perpendicular and parallel to the interface, a_{\perp} and a_{\parallel} , respectively, are determined by the symmetric (004) HRXRD rocking curves. a_{\parallel} is found to be in good agreement with the lattice constant of GaAs, and it was verified that all of the samples were grown as fully relaxed layer. The lattice constant of the relaxed GaAs_{1-x}N_x lattice, a_0 , is calculated giving to [16, 17]

$$a_0 = a_{\parallel} \left(1 - \frac{C_{11}}{C_{11} + 2C_{12}} \frac{a_{\parallel} - a_{\perp}}{a_{\parallel}} \right) \quad (1)$$

where C_{11} and C_{12} are the elastic constants of GaAs_{1-x}N_x, which are taken as the nitrogen compositions of weighted mean values between the values of GaAs and β -GaN. The elastic constants for GaAs_{1-x}N_x are calculated from the nitrogen composition using $C_{11} = 29.6 \times 10^{11}$, $C_{12} = 1.54 \times 10^{11}$ dyn/cm² for β -GaN and $C_{11} = 11.883 \times 10^{11}$, $C_{12} = 5.383 \times 10^{11}$ dyn/cm² for GaAs. The strain in the growth direction in the GaAs_{1-x}N_x strained layer on GaAs substrate is obtained as

$$\varepsilon_{\perp} = \frac{a_{\perp} - a_0}{a_0} \quad (2)$$

Also the in-plane strain in the GaAs_{1-x}N_x strained layer is obtained as

$$\varepsilon_{\parallel} = \frac{a_{\parallel} - a_0}{a_0} \quad (3)$$

Because a GaAs_{1-x}N_x strained layer grown on a GaAs (001) substrate is under biaxial stress, the in-plane strain ε_{\parallel} is related to ε_{\perp} by

$$\varepsilon_{\parallel} = -\frac{C_{11}}{C_{12}} \varepsilon_{\perp} \quad (4)$$

The nitrogen composition is determined assuming Vegard's rule,

$$a_{\text{GaAs}_{1-x}\text{N}_x} = xa_{\beta\text{-GaN}} + (1-x)a_{\text{GaAs}} - \theta_{\text{GaAsN}}(1-x)x \quad (5)$$

where a_{GaAs} and $a_{\beta\text{-GaN}}$ are the lattice constant of GaAs

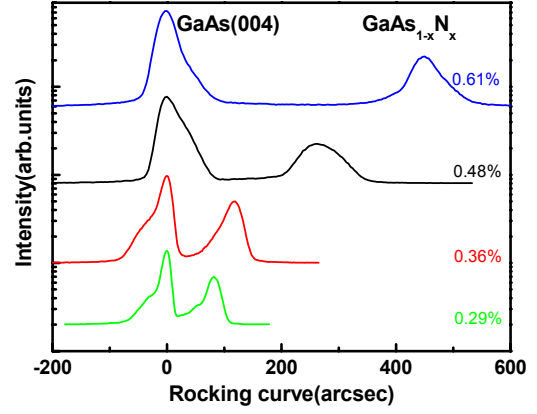


Fig. 1. HR-XRD (004) rocking curves of the strained GaAs_{1-x}N_x epilayers with of four different nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 %.

and β -GaN, respectively, θ_{GaAsN} is the lattice bowing parameter and calculated to be equal to -20.95 \AA [17]. From the standard (004) ω scans shown in Fig. 1, Bragg's law can be used quite easily to extract the in-plane lattice constants from the angular position $\omega_{\text{GaAsN}}^{004}$ of the peak associated with the strained GaAs_{1-x}N_x epilayers [15]. From the measured peak positions, the in-plane lattice constants are evaluated to be 5.62 \AA , 5.61 \AA , 5.60 \AA and 5.58 \AA , respectively, which are corresponding to the strained GaAs_{1-x}N_x epilayers for various nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 %. Those constants are also corresponding to in-plane tensile strains of 0.057 %, 0.071%, 0.094% and 0.119%, respectively. The in-plane tensile strain of the strained GaAs_{1-x}N_x epilayers increases as nitrogen composition increases.

The room-temperature Raman spectra were measured using a pumped solid-state laser unit of 532 nm to determine the strain percentage of the strained GaAs_{1-x}N_x epilayers. The spectra were taken at room temperature in a (001) backscattering geometry for the strained GaAs_{1-x}N_x epilayers with various nitrogen compositions. Fig. 2 shows the Raman spectra of the strained GaAs_{1-x}N_x epilayers with various nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 %. As can be seen from Fig. 2, the Raman peak shifts toward lower wave number with increasing nitrogen compositions, which indicates the presence of tensile strain in the strained GaAs_{1-x}N_x epilayers.

The Raman spectra of the strained GaAs_{1-x}N_x epilayers are observed to be dominated by the longitudinal optical

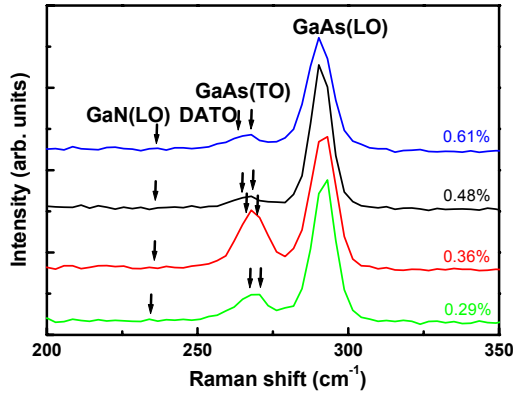


Fig. 2. Raman spectra of strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers with of four different nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 %.

(LO) phonon mode as the strongest peaks show up around $289\text{--}294\text{ cm}^{-1}$. Moreover, the broad and weak spectral features in the range of $255\text{--}276\text{ cm}^{-1}$ originate from the signals of transverse optical (TO) phonon mode, which is consistent with the observation of a disorder-activated transverse optical (DATO) phonon mode. The weak, broad feature located at $\sim 235\text{ cm}^{-1}$ can be assigned to the GaN-like LO mode, with possibly some small contribution of the forbidden GaN-like TO mode [18]. These two modes cannot be corrected because they are very close in frequency, as occurs in strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers for similar nitrogen compositions. On the other hand, the GaAs (LO) phonon peaks of strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers with various nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 % appear at 293.08 , 292.34 , 290.78 , and 289.11 cm^{-1} , respectively. The observed lower wave number shift in the phonon peak signifies that the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers are under tensile strain and nitrogen composition.

In theory, the measured shift of the GaAs(LO) peak can be expressed as

$$\Delta\omega = \Delta\omega_{\text{ally}} + \Delta\omega_{\text{strain}} \quad (6)$$

For biaxial strain, the magnitude of $\Delta\omega_{\text{strain}}$ in Eq. (6) depends linearly on the in-plane strain ϵ_{\parallel} as it follows the relationship [19]

$$\Delta\omega_{\text{strain}} = \frac{1}{\omega_0} \left(q - p \frac{C_{12}}{C_{11}} \right) \epsilon = b\epsilon_{\parallel} \quad (7)$$

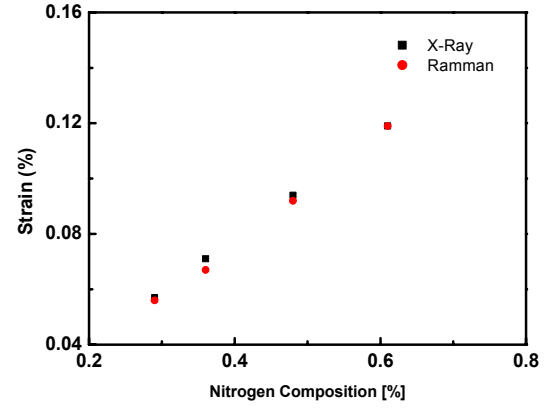


Fig. 3. Nitrogen compositions dependence of strain of four strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers with various nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 %, the strain was measured determined HR-XRD and Raman spectroscopy.

where ω_0 is the unstrained Raman frequency, C_{11} and C_{12} are elastic constants, and p and q are phonon deformation parameters. The magnitude of $\Delta\omega_{\text{alloy}}$ depends linearly on the nitrogen composition x as it follows the relationship

$$\Delta\omega_{\text{alloy}} = \omega_{\text{layer}} - \omega_{\text{wafer}} = ax \quad (8)$$

The in-plane tensile strain in the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayer is calculated from the strain versus Raman spectral peak shift ($\Delta\omega_{\text{strain}} = \omega_{\text{layer}} - \omega_{\text{wafer}}$) relative to the GaAs wafer. The equation can be simplified to $\Delta\omega_{\text{strain}} = b\epsilon_{\parallel}$, where ϵ_{\parallel} is the tensile strain and $b = -(62.7 \pm 4.0)\text{ cm}^{-1}$ [19]. The strain contribution $\Delta\omega_{\text{strain}}$ has to be deduced from the total Raman shift in order to determine the nitrogen composition dependence of the Raman shift. The Raman shift of relaxed $\text{GaAs}_{1-x}\text{N}_x$ alloys, $\Delta\omega_{\text{alloy}}$, can be easily calculated by using Eq. (8), where $a = -(7.54 \pm 4.0)\text{ cm}^{-1}$ [19]. The Raman shift of strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers, $\Delta\omega_{\text{alloy}}$, with various nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 % are evaluated to be 0.021 , 0.027 , 0.036 and 0.045 cm^{-1} , respectively. The estimated value of the Raman shift for strain of strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers for various nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 % are 293.08 , 292.34 , 290.78 , and 289.11 cm^{-1} , respectively. With Eq. (7), the in-plane tensile strains of strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers for various nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 % are of 0.056% , 0.067% , 0.092% and 0.119% , respectively. This result shows that the in-plane tensile strain of the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers

increases as the nitrogen composition increases. Fig. 3 show the nitrogen compositions dependence of strain of four strained GaAs_{1-x}N_x epilayers with various nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 %, the strain was measured determined HR-XRD and Raman spectroscopy.

The temperature dependences of the photocurrent spectra of strained GaAs_{1-x}N_x epilayers with various nitrogen compositions were measured at temperatures ranging from 300 to 30 K. Fig. 4 show the temperature dependence of photocurrent spectra of four strained GaAs_{1-x}N_x epilayers with various nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 %. The wavelength ranges from 600-1100 nm. The photocurrent spectra are dominated by the GaAs_{1-x}N_x-related transitions and the GaAs related band edge photocurrent peak seems to be completely dependent on the temperature, as can be clearly seen in Fig. 4. The GaAs_{1-x}N_x-related transition photocurrent spectra peaks are found to consist of two bands whose valence-band splitting increase with increasing nitrogen composition. The two features near the GaAs_{1-x}N_x-related transition photocurrent peaks in the low-energy peak of the photocurrent spectra of the strained GaAs_{1-x}N_x epilayers are related to the GaAs_{1-x}N_x fundamental energy gap. Epitaxial GaAs_{1-x}N_x epilayers grown on (100) GaAs substrate are under tensile strain. The doublet is the photocurrent peaks of the strained status of the GaAs_{1-x}N_x epilayers. We assign the first and the second photocurrent spectra peaks to the split light-hole (E_{LH}) and heavy-hole (E_{HH}) transitions in the strained GaAs_{1-x}N_x epilayers [21], respectively. This analysis provides us with fine information about the strain status and the composition of the strained GaAs_{1-x}N_x epilayers. The energy gaps associated with the transitions from E_{HH} and E_{LH} of valence bands to the conduction band, as well as the valence band splitting $\Delta E = E_{HH} - E_{LH}$, can be related to the in-plane strain.

Fig. 4 shows that the photocurrent peak wavelength was red shifted as increasing temperature due to the decreased band gap of the strained GaAs_{1-x}N_x epilayers.

All of the photocurrent spectra peaks are dominated by an intense optical band to band transition that is originated to transition of the valance band electrons. For photocurrent spectra, the low photon energy cut-off of the response spectra was determined by using the band gap energy or the absorption edge of the semiconductor.

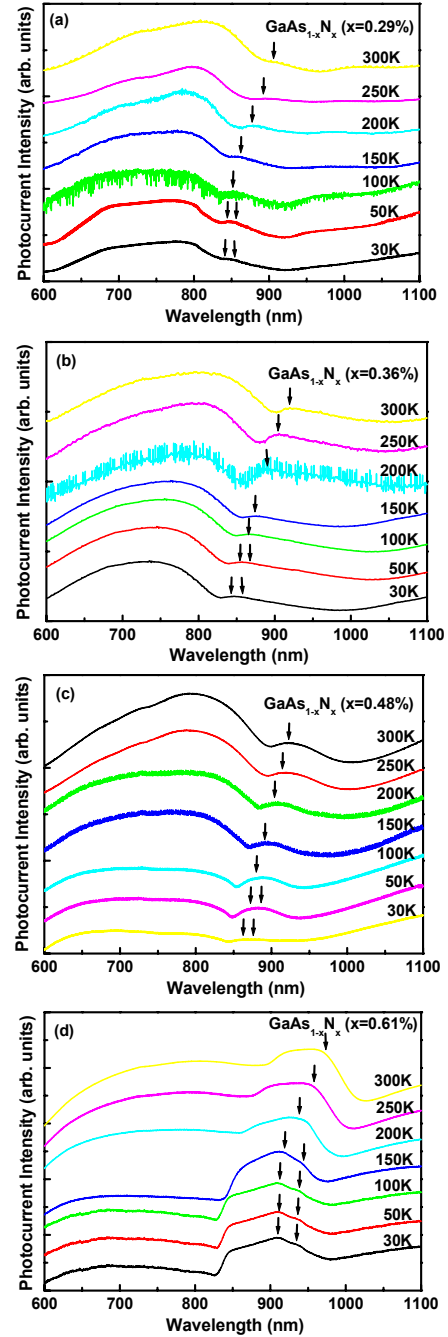


Fig. 4. Temperature dependence of the photocurrent spectra of the strained GaAs_{1-x}N_x epilayers with of four different nitrogen compositions of (a) 0.29 %, (b) 0.36 %, (c) 0.48 %, (d) 0.61 %.

A low photon energy cut-off was also observed in the response spectra characteristics because at low photon energy, the value of the absorption appear very large in semiconductors and all the incident optical band gap energy is absorbed near the surface. The photocurrent spectra show the valence band splitting peaks at low

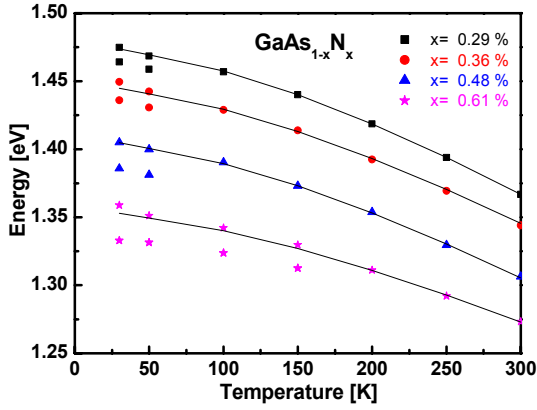


Fig. 5. Temperature dependence of the photocurrent peak energies of the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers with of four different nitrogen compositions of 0.29 %, 0.36 %, 0.48 % and 0.61 %.

temperature and their peaks are red-shifted with increasing temperature.

Fig. 5 summarizes the temperature dependence of the $\text{GaAs}_{1-x}\text{N}_x$ -related transition photocurrent peaks (E_{LH} and E_{HH}) energies of the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers with various nitrogen compositions. The temperature dependence of the direct energy band gap can be described by the Bose–Einstein expression proposed by Lautenschlager et al. [22] or the Varshni equation [23]. The Varshni relation, $E_g(T) = E_0 - \alpha T^2/(\beta + T)$, and the Bose–Einstein expression, $E_g(T) = E_B - a/[\exp(\theta/T) - 1]$, have been used to explain the temperature dependence of the fundamental band gap in semiconductors. E_g and E_B are the band gaps at $T = 0$ K while α and β are the Varshni coefficients. a is the electron-average phonon coupling constant, and θ corresponds to the average phonon temperature. Both expressions give similarly good fits to the measured $E_g(T)$ in GaAs. The parameters fitted for GaAs are $E_0 = 1.519$ eV, $\alpha = 5.405 \times 10^{-4}$ eV/K, and $\beta = 204$ K, and for GaN, $E_0 = 3.512$ eV, $\alpha = 5.66 \times 10^{-4}$ eV/K, and $\beta = 737.9$ K in the Varshni relation. For our experimental result, the temperature dependence of the $\text{GaAs}_{1-x}\text{N}_x$ -related transition photocurrent peaks (E_{LH} and E_{HH}) energies of the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayer can be described by using Varshni’s empirical expression. As shown in the Fig. 5, the solid lines are fits of the Varshni equations of the measured data for $\text{GaAs}_{1-x}\text{N}_x$ strained layers with nitrogen composition of 0.29 %, 0.36 %, 0.48 % and 0.61 %, respectively.

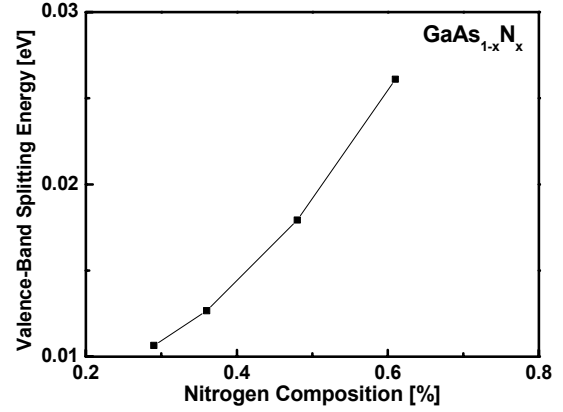


Fig. 6. Measured valence band splitting $\Delta E = E_{\text{HH}} - E_{\text{LH}}$ of strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers versus nitrogen composition dependence.

respectively. The fitted parameters for the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers with nitrogen composition show the expected trend of decreasing E_0 , α and β as increasing nitrogen composition. For the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers with nitrogen composition increase as shown in the Fig. 4 and 5, we observe a significant valence band heavy-holes to conduction and valence band light-holes to conduction transitions due to the strain-induced valence band splitting in the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers and due to the lattice mismatch to GaAs substrate. These photocurrent spectra with increasing nitrogen composition, the dominant valence band splitting $\Delta E = E_{\text{HH}} - E_{\text{LH}}$ to the increasing. The measured valence band splitting $\Delta E = E_{\text{HH}} - E_{\text{LH}}$ of the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers versus nitrogen composition dependence are shown in Fig. 6. As shown in the Fig. 6, the solid lines are fit the valence band splitting (ΔE) of the measured data for strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers with nitrogen composition and temperature dependence. The tensile strain is accompanied by splitting of light- and heavy-hole bands as the content of nitrogen composition increases to become 26.1 meV at $x=0.61\%$.

IV. CONCLUSIONS

We have investigated the Raman spectroscopy and the photocurrent spectra of the strained $\text{GaAs}_{1-x}\text{N}_x$ epilayers grown on GaAs (001) substrate by using metalorganic chemical vapor deposition with different nitrogen compositions. The nitrogen composition and strain in the

strained GaAs_{1-x}N_x epilayers was determined from both X-ray diffraction and Vegard's rule. The tensile strain of the strained GaAs_{1-x}N_x epilayers increases as the nitrogen composition increases. The Raman spectra are observed to be dominated by the longitudinal optical (LO) phonon mode as the strongest peaks show up around 287~292 cm⁻¹. Moreover, the weak and broad peaks features in the range of 265~270 cm⁻¹ originate from the peaks of transverse optical (TO) phonon mode. And the Raman peak shifts toward lower wave number with increasing nitrogen compositions, which indicates the presence of tensile strain in the strained GaAs_{1-x}N_x epilayers. The band gap and the valence-band splitting are obtained from the photocurrent spectra. As nitrogen composition increases, the tensile strain between the GaAs_{1-x}N_x epilayer and GaAs substrate increases while the valence-band splitting obtained from the photocurrent spectra increases.

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