A theoretical investigation of the band alignment of type-I direct band gap dilute nitride phosphide alloy of $GaN_xAs_yP_{1-x-y}$/GaP quantum wells on GaP substrates

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The GaP-based dilute nitride direct band gap material Ga(NAsP) is gaining importance due to the monolithic integration of laser diodes on Si microprocessors. The major advantage of this newly proposed laser material system is the small lattice mismatch between GaP and Si. However, the large threshold current density of these promising laser diodes on Si substrates shows that the carrier leakage plays an important role in Ga(NAsP)/GaP QW lasers. Therefore, it is necessary to investigate the band alignment in this laser material system. In this paper, we present a theoretical investigation to optimize the band alignment of type-I direct band gap $GaN_xAs_yP_{1-x-y}$/GaP QWs on GaP substrates. We examine the effect of nitrogen (N) concentration on the band offset ratios and band offset energies. We also provide a comparison of the band alignment of type-I direct band gap $GaN_xAs_yP_{1-x-y}$/GaP QWs with that of the $GaN_xAs_yP_{1-x-y}$/$Al_zGa_{1-z}P$ QWs on GaP substrates. Our theoretical calculations indicate that the incorporations of N into the well and Al into the barrier improve the band alignment compared to that of the GaAsP/GaP QW laser heterostructures.

Keywords: quantum well, band alignment, carrier confinement, dilute nitride phosphide alloy

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1. Introduction

In recent years, considerable physical and technical interest has been paid to the study of dilute nitride III–V alloys due to their potential optoelectronic device applications and unique N-induced physical properties.[1] These properties include a large band gap bowing and an increase in the electron effective mass[2] with only a few percent of incorporated nitrogen, which in turn enable telecommunication wavelength lasers to be grown on GaAs substrates and show the potential for more efficient lasers.[3] More recently, attention has turned to GaP based dilute nitrides where additional interesting phenomena and applications can also be explored.[4] Fortunately, there is a relatively small difference in the lattice constants of GaP and Si (less than 0.4% at room temperature), allowing the growth of GaP on Si without dislocations.[5] Although GaP is itself an indirect band gap semiconductor, the dilute nitride mixed compound Ga(NAsP) with a high As fraction exhibits a direct band gap material lattice matched to GaP and hence Si.[6] Lasing operation has been observed in these novel Ga(NAsP) material systems based on GaP,[7] and the first gain measurements emphasized that the material properties of these novel Ga(NAsP) material systems are comparable to the standard III/V material systems applied in the commercial laser diodes.[8] Hence, a Ga(NAsP)/GaP quantum well on a silicon substrate is a promising system for lasers.[9] Nevertheless, to optimize the design of an efficient laser structure, the heterojunction band offsets play an important role. To the best of our knowledge, the present study is the first that introduces a theoretical calculation of the band alignment of the quaternary $GaN_xAs_yP_{1-x-y}$ well material with GaP or AlGaP barriers on GaP and hence on Si substrates.

2. Theoretical models

2.1. Band anti-crossing model

We use the simple band anti-crossing model (BAC)[10,11] to exploit the concentration range in which we might expect a direct energy gap for dilute nitride mixed compound $GaN_xAs_yP_{1-x-y}$. The BAC model, based on the repulsion of a N-related level with the Γ level of the host material, has been widely used because of its simplicity and qualitative agreement with experimental observations, for example, the red shift of the band gap and the increase in the electron effective mass with increasing N concentration. The validity of the simple BAC model in describing GaP-based dilute nitride materials has been shown by Chamings *et al.*[4,12] According to the BAC model, the different behavior between GaAsN and GaPN can be explained by the difference of the nature of the host material band gap (direct for GaAs and indirect for GaP) and the position of the N-related level relative to the conduction band (CB) (above the CB of GaAs and below that of GaP).[13,14] This simple model describes the effect of nitrogen incorporation in dilute nitrides and allows us to calculate the fundamental band gap. In the BAC model, the interaction between a single localized N state and the extended conduction band states

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splits the CB into two subbands E_- and E_+. The energies of these two subbands can be calculated through

$$\begin{vmatrix} E-E_{\rm M} & -V_{\rm MN} \\ -V_{\rm MN} & E-E_{\rm N} \end{vmatrix} = 0, \quad (1)$$

where $E_{\rm M}$ and $E_{\rm N}$ are the energies of the extended state and of the localized N level relative to the top of the valence band, respectively. The interaction parameter $V_{\rm MN}$ is equal to $C_{\rm MN}\sqrt{x}$, where $C_{\rm MN}$ is the coupling parameter determined by the strength of coupling between the localized and the extended states, $C_{\rm MN}$ can be taken as 1.9 for $GaN_xAs_yP_{1-x-y}$,[12] x is the N concentration. The solution to this eigenvalue problem gives us the dispersion relation

$$E_\pm = \left(E_{\rm N}+E_{\rm M} \pm \sqrt{(E_{\rm N}-E_{\rm M})^2+4V_{\rm MN}^2}\right)/2, \quad (2)$$

where E_- describes the bulk band gap energy of the $GaN_xAs_yP_{1-x-y}$ alloy. The nitrogen dependence of the conduction band energy $E_{\rm M}$ of the matrix semiconductor can be taken as[15] $E_{\rm M} = E_0 - 3.5x$ for $GaN_xAs_yP_{1-x-y}$, where E_0 is the bulk band gap energy in the absence of nitrogen. For the band structures of the quaternary alloys $GaN_xAs_yP_{1-x-y}$/GaP, the band gaps are linearly interpolated from those of the binary materials using the following relations:[16]

$$\begin{aligned} T_{\rm GaNAs} &= uB_{\rm GaN} + (1-u)B_{\rm GaAs} + u(1-u)C_{\rm GaNAs}, \\ T_{\rm GaAsP} &= \upsilon B_{\rm GaAs} + (1-\upsilon)B_{\rm GaP} + \upsilon(1-\upsilon)C_{\rm GaAsP}, \\ T_{\rm GaNP} &= \omega B_{\rm GaN} + (1-\omega)B_{\rm GaP} + \omega(1-\omega)C_{\rm GaNP}, \end{aligned} \quad (3)$$

where $u = (1-x-y)/2$, $\upsilon = (2-x-2y)/2$, $\omega = (2-2x-y)/2$, and C is the bowing parameter for the ternary alloy. Finally, we calculate the bulk band gap energy E_0 of the ternary alloy taking into account the bowing parameters using the following expression:

$$\begin{aligned} &Q_{GaN_xAs_yP_{1-x-y}}(x,y) \\ &= \frac{xyT_{\rm GaNAs}(u)+y(1-x-y)T_{\rm GaAsP}(\upsilon)+x(1-x-y)T_{\rm GaNP}(\omega)}{xy+y(1-x-y)+x(1-x-y)}. \end{aligned} \quad (4)$$

The required material parameters for $GaN_xAs_yP_{1-x-y}$/GaP are tabulated in Tables 1 and 2.

Table 1. Binary material parameters used in calculations. The parameters are taken from Vurgaftman *et al.*,[17] except $E_{\rm v,av}$.[18]

Material	GaAs	GaN	GaP	AlP
Lattice constant a_0/A°	5.653	4.5	5.4505	5.4672
Energy gap E_0/eV	1.424	3.299	2.76	3.63
Spin–orbit splitting Δ_0/eV	0.34	0.017	0.08	0.07
Hydrostatic deformation potential for CB a_c/eV	−7.17	−6.71	−8.2	−5.7
Hydrostatic deformation potential for VB a_v/eV	−1.16	−0.69	−1.7	−3.0
Elastic stiffness constant C_{11}/GPa	1221.0	293.0	1405.0	1330.0
Elastic stiffness constant C_{12}/GPa	566.0	159.0	620.3	630.0
Deformation potential b/eV	−2.0	−2.0	−1.6	−1.5
$E_{\rm v,av}$	−6.92	–	−7.40	−8.09

Table 2. Bowing parameters used in the calculation of E_0, $E_{\rm g}^X$, and $E_{\rm g}^L$ of the related III–V ternaries.[16]

	E_0	$E_{\rm g}^X$	$E_{\rm g}^L$
$C_{\rm AlGaP}$	0	−0.13	–
$C_{\rm GaNP}$	−3.9	–	–
$C_{\rm GaNAs}$	$-120.4+100x$	–	–
$C_{\rm GaAsP}$	−0.19	–	–

2.2. Model solid theory

It has been shown previously that the band alignment of GaInNAs on GaAs and InP substrates can be analyzed by means of the model solid theory ignoring the presence of nitrogen in the average valence band energy and taking into account the presence of nitrogen in all other laser parameters.[19–21] The calculated results were in agreement with the experimental data.[21] Therefore, we use the model solid theory to calculate the band alignment of GaNAsP alloys on GaP substrates.

The relative band alignment of the band edges between the quantum well and the barrier is the total band discontinuity distributed over the conduction and the valence bands, $\Delta E_{\rm C}$ and $\Delta E_{\rm V}$, respectively. The band discontinuity depends on the semiconductors and the amount of mismatch strain at the interface. It has been commonly accepted that the nitrogen incorporation mainly affects the conduction band states of the dilute nitride III–V alloys, leading to an increase in the conduction band offset $Q_{\rm C}$ and a small discontinuity in the valence band edge.[22] According to Van de Walle's model solid theory,[23] the band offset ratio for conduction and valence bands, $Q_{\rm C,V}$, is determined by the discontinuity fractions of $\Delta E_{\rm C,V}/\Delta E_{\rm g}$. The energy of the potential barrier, $\Delta E_{\rm g}$, is determined from the difference between the bulk band gap energy of the barrier layers and the strained band gap energy of the active layer. The effects of the strain in the plane of the epitaxial growth are calculated as

$$\varepsilon = (a_{\rm s}-a_{\rm e})/a_{\rm e}, \quad (5)$$

where $a_{\rm s}$ is the lattice constant of the substrate, and $a_{\rm e}$ is the lattice constant of the quaternary epitaxial layer.[24] The conduction band position can be calculated by simply adding the

strained band gap energy to the valence band position. The unstrained valence band edge of the active region material is set as the reference energy of zero. The valence band position is given by

$$E_{\rm v}(x,y)=\begin{cases}E_{\rm v,av}(x,y)+\dfrac{\Delta_0(x,y)}{3}+\delta E_{\rm hh}(x,y),\\ \qquad\text{for hh (compressive strain)},\\ E_{\rm v,av}(x,y)+\dfrac{\Delta_0(x,y)}{3}+\delta E_{\rm lh}(x,y),\\ \qquad\text{for lh (tensile strain)},\end{cases}\tag{6}$$

where $E_{\rm v,av}(x,y)$ is the average valance subband energy, and Δ_0 is the spin–orbit band energy.[25] These values are listed in Table 1. The conduction band is shifted by the energy $\delta E_{\rm c}(x,y)$

$$\delta E_{\rm c}(x,y)=2a_{\rm c}\left(1-\frac{C_{12}}{C_{11}}\right)\varepsilon.\tag{7}$$

The valance bands are shifted by the energy $\delta E_{\rm hh}(x,y)$ and $\delta E_{\rm lh}(x,y)$

$$\begin{aligned}\delta E_{\rm hh}(x,y)&=-P_\varepsilon-Q_\varepsilon,\\ \delta E_{\rm lh}(x,y)&=-P_\varepsilon+Q_\varepsilon,\end{aligned}\tag{8}$$

with

$$\begin{aligned}P_\varepsilon&=-2a_{\rm v}\left(1-\frac{C_{12}}{C_{11}}\right)\varepsilon,\\ Q_\varepsilon&=-b\left(1+\frac{2C_{12}}{C_{11}}\right)\varepsilon,\end{aligned}\tag{9}$$

where $a_{\rm c}$ and $a_{\rm v}$ are the conduction- and valance-band hydrostatic deformation potentials, b is the valance band shear deformation potential, and C_{11} and C_{12} are the elastic stiffness constants.[25] The strained band gaps can be expressed as[25]

$$\begin{aligned}E_{\rm c-hh}(x,y)&=E_{\rm g}(x,y)+\delta E_{\rm c}(x,y)-E_{\rm hh}(x,y),\\ E_{\rm c-lh}(x,y)&=E_{\rm g}(x,y)+\delta E_{\rm c}(x,y)-E_{\rm lh}(x,y).\end{aligned}\tag{10}$$

The conduction band position is

$$E_{\rm c}(x,y)=\begin{cases}E_{\rm v}(x,y)+E_{\rm c-hh}(x,y),\\ \qquad\text{for hh (compressive strain)},\\ E_{\rm v}(x,y)+E_{\rm c-lh}(x,y),\\ \qquad\text{for lh (tensile strain)},\end{cases}\tag{11}$$

$$\frac{\Delta E_{\rm c}}{\Delta E_{\rm g}}=1-\frac{E_{\rm v}^{\rm w}-E_{\rm v}^{\rm b}}{E_{\rm g}^{\rm b}-E_{\rm g}^{\rm w}},\tag{12}$$

where $E_{\rm v}^{\rm w}$ and $E_{\rm v}^{\rm b}$ are the valance band positions in the well and the barrier materials, respectively, and $E_{\rm g}^{\rm w}$ and $E_{\rm g}^{\rm b}$ are the strain adjusted band gaps ($E_{\rm c-hh}$ for the compressive strain and $E_{\rm c-lh}$ for the tensile strain) for the well and the barrier materials.[25]

3. Results and discussion

It has been stated by Chamings *et al.*[12] that the carrier leakage plays a significant role in $GaN_xAs_yP_{1-x-y}$/GaP QW lasers. They claimed that if these leakage paths can be reduced, the growth of a direct band gap material on GaP and eventually Si substrates may lead to fully monolithic silicon-based optoelectronic integrated circuits (OEICs) operating at room temperature. The optimization of the band alignment of the $GaN_xAs_yP_{1-x-y}$/ GaP QW structure on a GaP substrate plays an important role to reduce the carrier leakage to the barriers. The present study introduces, for the first time, a theoretical calculation of the band offset ratio and the band offset energies for conduction and valence bands by Van de Walle's model solid theory to achieve our goal.

We aim to present the calculations of the strained band gap in these newly proposed compressively strained $GaN_xAs_yP_{1-x-y}$/GaP type-I QWs on Si substrates. To achieve this, we have to determine the concentration range in which the quaternary $GaN_xAs_yP_{1-x-y}$ alloy behaves like a direct band gap semiconductor. Thus, we first determine the concentration range in which we expect a direct energy gap of ternary $GaAs_yP_{1-y}$.

Figure 1 shows the calculated variations of energy levels at Γ, X, and L minimums of the conduction band of the ternary material $GaAs_yP_{1-y}$ with the phosphorus concentration. All of these energies are defined with respect to the zone center valence band (VB) maximum. As can be seen from Fig. 1, all the energy levels of Γ, X, and L minimums of the conduction band increase with increasing phosphorus concentration and Γ and X minimums of the conduction band crossover at the phosphorus mole fractions of about 45%. This indicates that $GaAs_yP_{1-y}$ has an indirect band gap above 45%. Therefore, we should prefer As-rich $GaAs_yP_{1-y}$ ternary materials to maintain in the direct band gap region.

Figure 1 presents the position of the nitrogen energy level $E_{\rm N}$, which has been determined by an interpolation of the isolated N level in ternary composition end points of GaAs:N and GaP:N.[15] The lower part of Fig. 1 indicates the calculated variation of bulk direct energy band gap E_- and its strained value $E_{\rm str}$ for the quaternary material $GaN_{0.04}As_yP_{0.96-y}$ alloy system as a function of the phosphorus concentration using the band anti-crossing model taking into account the macroscopic strain. The comparison of the upper and the lower parts of Fig. 1 indicates that the incorporation of nitrogen into $GaAs_yP_{1-y}$ causes a strong reduction in the band gap energy of the dilute nitride phosphide $GaN_{0.04}As_yP_{0.96-y}$ alloy. The nitrogen incorporation into $GaAs_yP_{1-y}$ also allows the required adjustment of the lattice constant to that of GaP.

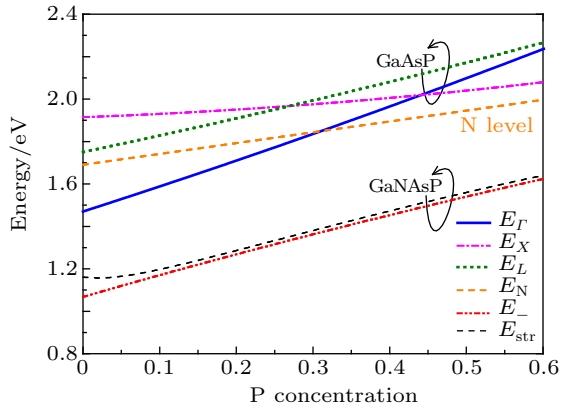


Fig. 1. (color online) Calculated energy positions of Γ, X and L minimums of the conduction band of the ternary $GaAs_yP_{1-y}$ alloy system and the fundamental band gap E_- and the strained band gap E_{str} of the quaternary $GaN_{0.04}As_yP_{0.96-y}$ on GaP as a function of the P concentration.

In order to investigate the N dependence of the direct band gap As-rich $GaN_xAs_yP_{1-x-y}$ alloy system, we choose an As concentration of 80%. As illustrated in Fig. 2, both bulk direct energy band gap E_- and its strained value E_{str} of $GaN_xAs_{0.8}P_{0.2-x}$ decrease rapidly as a function of the nitrogen concentration. This is due to the fact that there is a stronger repulsion between the extended state E_M and the localized N level E_N, leading to a red shift in the strained band gap with increasing N incorporation. These calculations confirm the possibility of the emission wavelength of the novel As rich $GaN_xAs_yP_{1-x-y}$/GaP material system being pushed beyond 1100 nm, possibly allowing laser devices with emission energies below the indirect band gap of Si at 1.124 eV.[26]

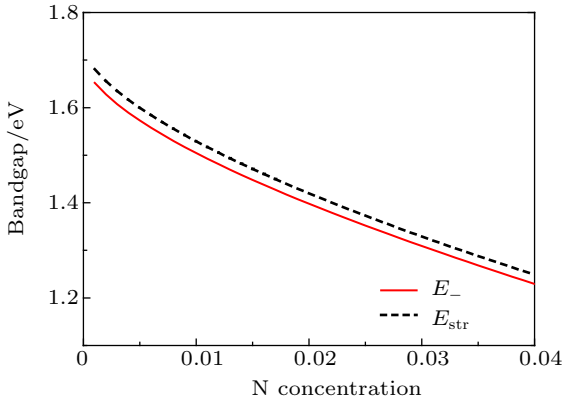


Fig. 2. The calculated variation of bulk direct energy band gap E_- and its strained value E_{str} for $GaN_xAs_{0.8}P_{0.2-x}$ on GaP with increasing nitrogen concentration.

We now extend our analysis to estimate the band alignment in these newly proposed As rich $GaN_xAs_yP_{1-x-y}$/GaP on GaP substrates using the model solid theory. Figure 3(a) presents the calculated band offset ratios of Q_c and Q_v for the compressively strained As-rich $GaN_xAs_{0.8}P_{0.2-x}$ type-I quantum well with GaP barriers on GaP substrates. Upper and lower curves represent the variations of the conduction band offset ratio Q_c and the valence band offset ratio Q_v with increasing nitrogen concentration in the well, respectively. These calculations show that the incorporation of N into $GaAs_yP_{1-y}$ increases the conduction band offset ratio Q_c and decreases the valence band offset ratio Q_v gradually. The corresponding band offset energies for conduction band ΔE_C and valence band ΔE_V are shown in Fig. 3(b). It can be seen from Fig. 3(b) that larger conduction band offset energies can be achieved with an increase in the nitrogen concentration of $GaN_xAs_{0.8}P_{0.2-x}$. The larger conduction band offset energies, in turn, cause an improved electron confinement in the well and a decreased electron spill-out. It can be concluded from Fig. 3 that the incorporation of nitrogen into $GaAs_yP_{1-y}$ mainly affects the conduction band offset energy and there are only minor changes in the valence band offset energy. Hence, we illustrate that the incorporation of nitrogen into $GaAs_yP_{1-y}$ leads to the N-containing system having a band alignment of that of the ideal case (a deep conduction well and a shallow valence well).

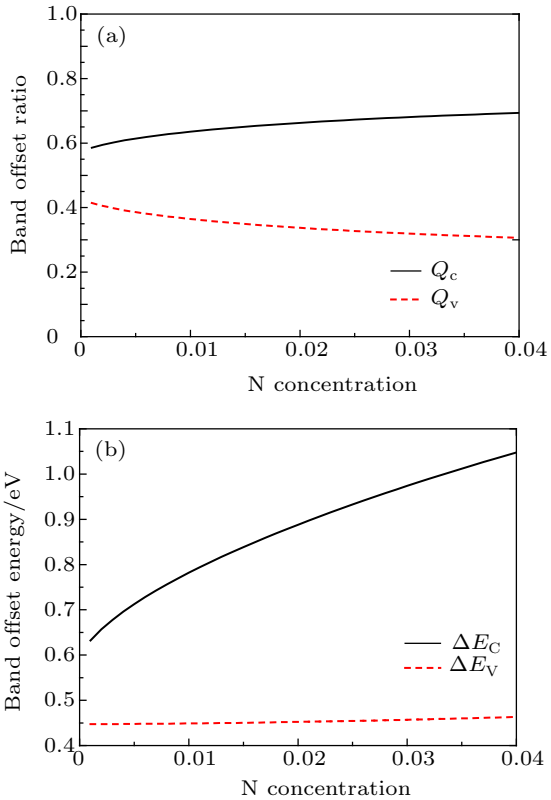


Fig. 3. The variations of (a) band offset ratios Q_c and Q_v and (b) band offset energies ΔE_C and ΔE_V in $GaN_xAs_{0.8}P_{0.2-x}$/GaP QW structure on GaP substrate as a function of nitrogen concentration.

Finally, we aim to reveal whether the optimization of the quaternary $GaN_xAs_yP_{1-x-y}$ well material on GaP substrates can be achieved by means of using $Al_zGa_{1-z}P$ barriers. Figure 4(a) presents the effect of the aluminium concentration in the barrier on the calculated band offset ratios Q_c and Q_v for the compressively strained type-I $GaN_xAs_{0.8}P_{0.2-x}/Al_zGa_{1-z}P$

QW structure on a GaP substrate for different N concentrations. It is seen from Fig. 4(a) that valence band offset ratio Q_v increases and conduction band offset ratio Q_c decreases with increasing Al concentration. However, it is clear from Fig. 4(b) that the combined effect of the N concentration in the well and the Al concentration in the barrier improves both band offset energies of conduction band ΔE_C and valence band ΔE_V. Thus, replacing GaP barriers with $Al_zGa_{1-z}P$ barriers causes a further increase at the depths of the conduction and the valence wells, leading to better carrier confinement in both conduction and valence bands.

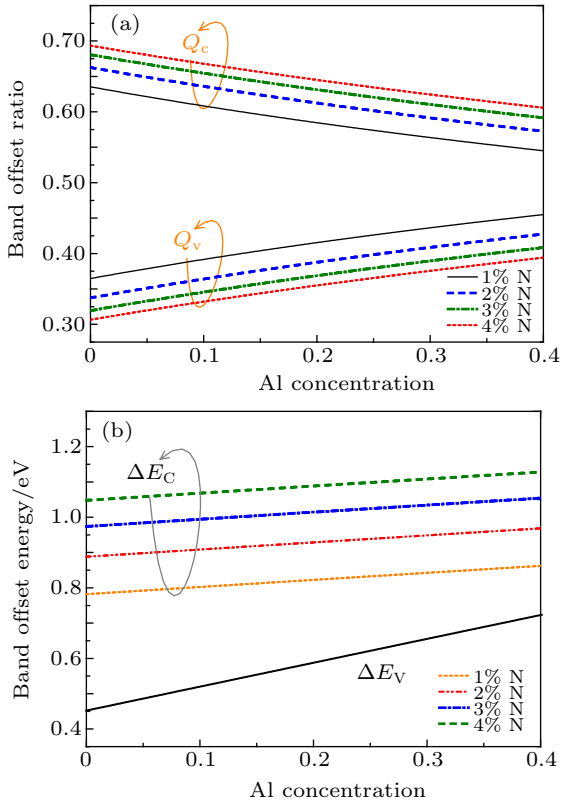


Fig. 4. (color online) Effect of N concentration (in well) and Al concentration (in barrier) on (a) band offset ratios Q_c and Q_v and (b) band offset energies ΔE_C and ΔE_V in $GaN_xAs_{0.8}P_{0.2-x}/Al_xGa_{1-x}P$ on GaP substrates.

4. Conclusion

We present a theoretical analysis to investigate and optimize the band alignment of the newly proposed $GaN_xAs_yP_{1-x-y}$/GaP QW on GaP substrates. Our results demonstrate that the band offset energy of the conduction band of the dilute nitride phosphide alloy increases rapidly with an increase in the nitrogen concentration. We have shown that the use of an $Al_zGa_{1-z}P$ barrier instead of a GaP barrier improves the band alignment, bringing an advantage to the novel quaternary dilute nitride laser system of $GaN_xAs_yP_{1-x-y}/Al_zGa_{1-z}P$ on GaP and hence Si substrates.

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