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RESEARCH ARTICLE

Effect of indium distribution on optical properties in InGaAs/GaAs quantum wells

Guozhi JIA (✉)[1], Jianghong YAO[2], Yongchun SHU[2], Xiaodong XIN[2], Biao PI[2]

1 Tianjin Institute of Urban Construction, Tianjin 300384, China
2 The Key Lab of Advanced Technique and Fabrication for Weak-Light Nonlinear Photonics Materials, TEDA Applied Physics School, Nankai University, Tianjin 300475, China

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Abstract The effect of In surface segregation and diffusion on the transition energy of an InGaAs/GaAs strained quantum well (QW) was investigated theoretically. Diffusion equations and the Schrödinger equation on the InGaAs/GaAs QW were solved numerically. The energy shifts under different diffusion lengths were simulated. When the width of QW, *L*, is larger than 5 nm, the change rate of the transition energy is very minimal at the initial stage of the annealing process for wide QW, and the transition energy has a rapid blue shift with an increase of the diffusion length. When *L* is smaller than 5 nm, the transition energy is very sensitive to the diffusion length. The change rate of transition energy increases with a decrease in QW width.

Keywords quantum well (QW), InGaAs/GaAs, surface segregation, diffusion

1 Introduction

In the last few years, the InGaAs/GaAs heterostructure system has received increasing interest from the electronic device and microwave industries. The strained quantum well (QW) laser has a more beneficial performance than other QW lasers such as lower threshold current density [1] and wider modulation bandwidth [2], and the emission wavelength of the strained QW laser can be adjusted by appropriately changing the strain [3]. With regard to the InGaAs/GaAs material system, In distribution directly decides the performances of devices. There are two major processes that influence In distribution in the InGaAs/GaAs heterostructure with the molecular beam epitaxy technology: surface segregation in the growth process [4] and interdiffusion in the annealing process [3].

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E-mail: jiaguozhi@tjuci.edu.cn

The heteroepitaxy of InGaAs layers on GaAs substrates is characterized by a strong segregation of indium atoms which accumulate at the growth front. This substantially modifies the In-composition profile, resulting in different electronic and optical properties of the devices based on that material. Segregation determines the composition profiles of QW in the growth direction in many semiconductor systems and consequently, the quantized levels are strongly influenced [4]. Some efforts have been achieved to minimize In segregation by controlling the growth temperature. However, growth at low temperature often results in poorer crystalline quality and usually has a negative influence on the final performance of the device [4,5]. On the other hand, In surface segregation leads to the appearance of the In-rich regions between the InGaAs and GaAs top barrier layer due to trapping of free excitons in the QW [6,7]. Thus, it is very important to analyze the effect of segregation on the composition distribution. Meanwhile, interdiffusion of atoms across the QW heterojunction plays an important role in tailoring the compositional and confinement profiles. In addition, more investigations have been made to analyze interdiffusion and tune the emission wavelength accurately [3]. An asymmetrical In profile is induced by the surface segregation, which also makes it troublesome to control the emission wavelength through the annealing process. Interdiffusion is a thermal process that facilitates the movement of constituent atoms across the heterojunction to eliminate the defect, dislocation and strain relaxation. Although extensive papers have been published to discuss and analyze the surface segregation [4,7–13], controlling or utilizing segregation to obtain an abrupt heterojunction still remains a challenge.

Based on previous work, this study was made to take the effects of surface segregation, growth mode and interdiffusion on the QW potential profile into account. In segregation processes based on the Muraki model are illustrated in detail. The effect of segregation efficiencies on

photoluminescence (PL) peak energy for $In_{0.25}Ga_{0.75}As$/GaAs with varied QW width and effect of annealing time and temperature on In distribution are also investigated.

2 Theoretical model

2.1 Surface segregation

The Muraki model adopted in this research can be described as follows: it is assumed that a certain fraction R of indium atoms on the topmost layer segregates to the next layer, and the remaining 1–R of In atoms are incorporated into the bulk before the next monolayer is completed. It has been proven by experiment that the model can reflect the growth progress perfectly [4]. According to the Muraki model, In composition in the nth layer is given by Refs. [,14] as

$$\text{for QW, } x(n) = x_0(1-R^n),\ 0 \leqslant n \leqslant N; \quad (1)$$

$$\text{for barrier, } x(n) = x_0(1-R^N)R^{n-N},\ n \geqslant N, \quad (2)$$

where x_0 and N are the nominal In composition and the well width in monolayer (ML), respectively, R is denoted as the segregation efficiency and growth of the Muraki starts at the ML $n = 0$. Consequently, In concentration in the surface layer (ML $n + 1$) is given by Refs. [,14] as

$$x_{\text{surf}}^{\text{In}}(n+1) = \frac{R}{1-R} x_{\text{bulk}}^{\text{In}}(n). \quad (3)$$

2.2 Diffusion model

The In composition profile of an InGaAs/GaAs QW after interdiffusion caused by annealing can be obtained by solving the conventional diffusion equation [15] as

$$\frac{\partial}{\partial t} C_{\text{In}}(x,t) = D(T)\frac{\partial^2}{\partial x^2} C_{\text{In}}(x,t), \quad (4)$$

and Eq. (4) can be solved and expressed as [15]

$$\chi(x) = \frac{1}{2}\chi_0\left[\text{erf}\left(\frac{h-x}{2\sqrt{Dt}}\right) + \text{erf}\left(\frac{h+x}{2\sqrt{Dt}}\right)\right], \quad (5)$$

where χ is the In mole fraction at position x, h is the half-width of the InGaAs QW , χ_0 is the In mole fraction of the as-grown InGaAs layer, D is the unknown intrinsic In-Ga interdiffusion diffusivity, and t is annealing time.

2.3 Transition energy

The emission photon energy of $In_xGa_{1-x}As$/GaAs QW can be written as

$$E_{\text{total}} = E_g^0(x) + \Delta E_{\text{strain}} + \delta E(x,L), \quad (6)$$

where $E_g^0(x)$ is the unstrained $In_xGa_{1-x}As$ band gap, ΔE_{strain} is the strain-induced change of the heavy-hole bands, and L is the width of QW. $\delta E(x, L)$ is the quantized energies for electrons or for holes, and it can be obtained by solving the Schrödinger equation numerically.

The band-to-band transition energy for the unstrained $In_xGa_{1-x}As$ layer at 300 K is related to the In mole fraction x as

$$E_g^0 = 1.424 - 1.614x + 0.54x^2. \quad (7)$$

The band gap shift caused by strain is assumed to linearly depend on the compressive biaxial strain, given by Refs. [15,16] as

$$\Delta E_{\text{strain}} = \left\{2a\left[\frac{C_{11}-C_{12}}{C_{11}}\right] - b\left[\frac{C_{11}+2C_{12}}{C_{11}}\right]\right\}\varepsilon, \quad (8)$$

where C_{11} and C_{12} are the elastic stiffness constants; a and b are the hydrostatic and shear deformation potentials, respectively; ε is the in-plane strain; $\varepsilon = (a_w - a_s)/a_s$, a_w is crystal lattice constant of the epitaxy layer; and a_s is crystal lattice constant of the substrate.

To obtain the quantized energy in QW and better understand the effect of In distribution on the optical properties of InGaAs/GaAs strained QW after annealing, a confining potential is essential to describe the InGaAs/GaAs QW. Gonzalez presented a simple formula [16] described as

$$V_0(z) = -\frac{U(x)}{\cosh^2(z/L)}, \quad (9)$$

where $U(x)$ represents the height of the barriers of the QW with thickness L, In nominal concentration x, z denotes the length along the crystal growth direction, and the QW is centered at $z = 0$. Although there is experimental evidence indicating that the potential profile with surface segregation in InGaAs/GaAs structures cannot be modeled with sufficient accuracy by a symmetric well, as given by Eq. (9) [16], it can be still proper to describe the potential profile based on the two-step growth process after annealing in this paper.

The values for the electron energy and hole energy can be obtained from the one-dimensional Schrödinger equation as [16]

$$E_n = -\frac{\hbar^2(1/L)^2}{8m_e^*}\left(-1+\sqrt{1+\frac{8m_e^* V_0}{\hbar^2 L^2}}\right)^2 - \frac{\hbar^2(1/L)^2}{8m_h^*}\left[-1+\sqrt{1+\frac{8m_h^* V_0}{\hbar^2 L^2}}\right]^2, \quad n = 1,2,3,\ldots, \quad (10)$$

where m_e^* and m_h^* are the electron and hole effective mass of the carrier in the well, respectively; V_0 is the depth of QW (equal total band offset energy); E_n is the quantized energy levels within the conduction bands or valence band; and $\hbar$ is the reduced Planck constant (i.e., $h/2\pi$). The transition energy from the first quantized electron state to the first heavy hole state can be then calculated. The 67:33 band offset ratio is assumed to calculate the conduction band and the valence band profiles. The values of all parameters for InGaAs are shown in Table 1.

Table 1 Room temperature material parameters as a function of In fraction used in numerical calculation (m_0 is the rest mass of free electrons)

quantity	variation with x
m_e/m_0	$0.075-0.0475x$
m_h/m_0	$0.55m_0$
a/eV	$-(8.67-5.66x)$
b/eV	$1.7+0.1x$
$C_{11}/10^2$ N·m^{-2}	$12.25-3.61x$
$C_{12}/10^2$ N·m^{-2}	$5.7-0.85x$
$a_s/10^{-1}$nm	5.6533
$a_w/10^{-1}$nm	$5.6533+0.4051x$

3 Results and discussion

Figure 1 shows the simulated concentration profiles of $In_{0.25}Ga_{0.75}As$/GaAs QW for different segregation efficiencies based on the Muraki model. Here, it should be pointed out that: 1) the profile of the incorporated In is an asymmetrical shape, which broadens In concentration profiles; 2) the In concentration profile is a gradual composition in the InGaAs layer; 3) In concentration becomes higher as the InGaAs thickness increases. All the results above match well with the experimental results [7,17]. Generally speaking, In segregation to the growth surface could result in a gradual composition of the interface and an In surface enrichment, which influences the optical properties of QW.

To control the gradual composition within a very small range, and to keep In content constant in QW, the corresponding critical thickness of the InGaAs layer on GaAs substrate should be determined by the theory of Matthews and Blakeslee [18]. In content will be constant in QW if the nominal In content deposited in the second step satisfies the condition

$$x(n)_{\text{bulk}} = [x(n)_{\text{surf}} + x(n+1)_{\text{nominal}}](1-R), \tag{11}$$

where $x(n)_{\text{surf}}$ and $x(n)_{\text{bulk}}$ are the concentrations of the nth in the surface and bulk phases, respectively.

In fact, it is well known that composition concentration can be varied by the ratio of group III element fluxes, so as the thickness of the well by the duration of the deposition and segregation efficiency by the growth temperature or the V/III ratio [4,14]. This indicates that an approximately abrupt heterojunction can be realized in experiments according to proper growth conditions. Previously, the growth rate should be very low to decrease the effect of segregation, while the interdiffusion process is mainly promoted by the diffusion of Ga vacancies in the GaAs-based heterostructure system. The strain in the materials would also accelerate the interdiffusion process.

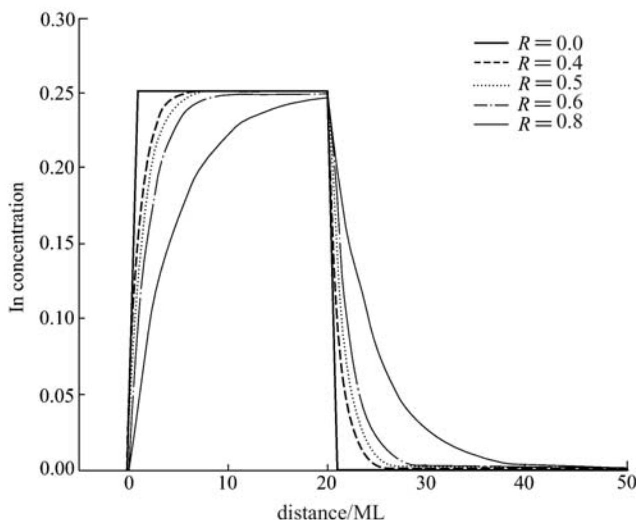

Fig. 1 Simulated In concentration profiles for different segregation efficiencies R, assuming a QW thickness $N = 20$ ML and a nominal composition of $x_0 = 0.25$

The annealing process is essential to obtain a high quality semiconductor film material and device application, and the major aim is to eliminate the strain relaxation, defect and other unwanted variables. In addition, interdiffusion is also important in tuning the emission wavelength used in standard dense wavelength division multiplexed (DWDM) optical communication systems [3]. Figure 2 shows the In concentration profiles of $In_{0.25}Ga_{0.75}As$/GaAs QW with a well width of $L = 6.0$ nm for different diffusion lengths L_d. The as-grown square well structure gradually changes from an abrupt interface to a graded profile as a result of the interdiffusion of atoms. Thus, a confining potential Eq. (9) used in the transition energy simulation is practical in the annealing sample. The transition energy of InGaAs/GaAs interdiffused QW as a function of diffusion length is shown in Fig. 3. The annealing process can influence the In composition profile and further influence the transition energy due to the change of compositional alloys, strain, and subband energy of the InGaAs/GaAs QW. It is worth noting that when L is larger than 5 nm, the change rate of the transition energy is very minimal at the initial stage of the annealing process for wide QW, and the transition energy has a rapid blue shift with an increase of the diffusion length. When L is smaller than 5 nm, the transition energy

becomes very sensitive to the diffusion length, and the change rate of transition energy increases with decreasing QW width. The diffusion length is defined as $L_d = \sqrt{Dt}$, where D can be expressed as $D_0\exp(-E/k_BT)$. k_B is Boltzmann constant, T is the temperature for the annealing process, D_0 is the diffusion constant, and E is the activated energy of In-Ga interdiffusion describing the temperature dependence of D [19,20]. Thus, the transition energy can be modulated by changing the annealing temperature and time after obtaining the activated energy of interdiffusion. It is essential to choose the appropriate In content and width of QW to tune the emission wavelength accurately through the annealing process.

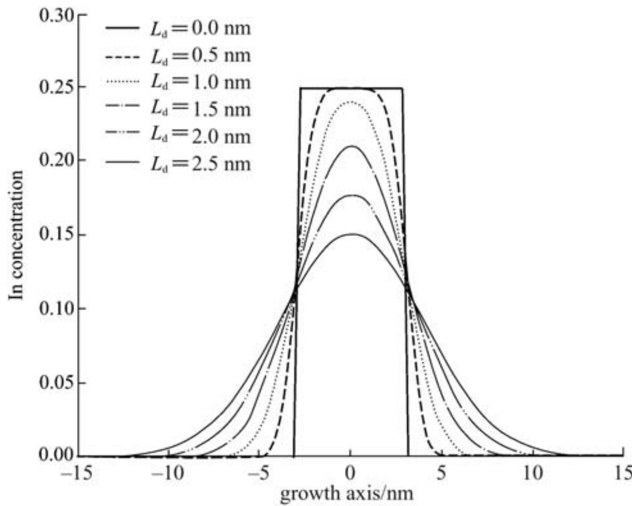

Fig. 2 Simulated In concentration profiles of $In_{0.25}Ga_{0.75}As$/GaAs QW as a function of diffusion length, assuming a well width L = 6.0 nm

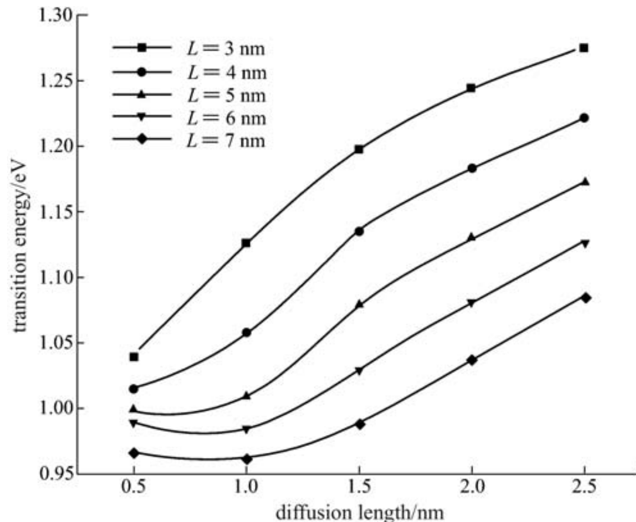

Fig. 3 Transition energy of $In_{0.25}Ga_{0.75}As$/GaAs interdiffused QW as a function of diffusion length for different QW widths

4 Conclusion

The effects of segregation and diffusion on the transition energy of InGaAs/GaAs strained QW have been investigated. The theoretical simulation results indicate that there is a critical width of about 5 nm for $In_{0.25}Ga_{0.75}As$/GaAs QW. When L is smaller than 5 nm, the transition energy becomes very sensitive to the diffusion length.

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