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Determination of diffusion lengths for intermixed quaternary quantum well with polarized edge-emitting photoluminescence

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Diffusion lengths of the group III and V sublattices are quantitatively determined for intermixing of a quaternary InGaAs/InP quantum well using polarized edge-emitting photoluminescence. Diffusion-length loci are plotted on a contour diagram of wavelength shifts of electron-heavy-hole and electron-light-hole transitions and illustrate the time progression of intermixing clearly. The plasma-enhanced intermixing effect is investigated using this technique, showing the difference of intermixing development in the early and the late stages. It is seen that the calculated diffusion-length ratio may change with the annealing duration. © 2007 American Institute of Physics.

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For decades quantum-well intermixing (QWI) has attracted considerable interest in the application of monolithic integration of quantum well (QW) based optoelectronic devices. Intermixing in a quaternary material system, e.g., InGaAs(P)/InP, is more complicated than that in a ternary material system such as AlGaAs/GaAs, because interdiffusion occurs on both group V and III sublattices and has to be characterized by two parameters: the diffusion lengths of both sublattices L_V and L_{III} . Unless a diffusion-length ratio, i.e., $k=L_V/L_{III}$, is presumed (see Ref. 1, for example), a conventional photoluminescence (PL) measurement cannot reveal the diffusion lengths and thus diffusion coefficients. On the other hand, it is important to determine k values experimentally in a process development of QWI because it plays an important role in controlling the band gap of quaternary QWs. k must be greater than 1 if a large blueshift is anticipated, whereas a redshift can result from a sufficiently small k .^{2,3} The former has an application in polarization insensitive devices⁴ and the latter can be used to realize low loss and mode filtering devices.⁵ Strain may also result from a non-unit diffusion-length ratio near well-barrier interfaces. Several methods, such as scanning tunneling microscopy,⁶ double crystal x-ray diffractometry,⁷ high-resolution x-ray diffraction,⁸ differential reflectance,⁹ and absorbance measurement,¹⁰ were used to estimate the diffusion-length ratio. However, these experiments either exhaust time, damage samples, or demand large sample size, and thus are not suitable for investigations which involve large volume of samples and tests.

Since the electron-heavy-hole (C-HH) and electron-light-hole (C-LH) transitions contribute to the TE- and TM-polarized photoluminescence (PPL) in different shares, detection of the PPL spectra from edge emission allows us to obtain more details of the electronic energy structure. From PPL spectra, certain material characteristics can be determined, e.g., strain in bulk and QW materials.^{11,12} In this letter, we report an experimental technique to determine the

diffusion lengths L_V and L_{III} in a quaternary InGaAs/InP QW based on measurement of room-temperature edge-emitting PPL spectra.

Samples used in the present investigation have a lattice-matched In_{0.53}Ga_{0.47}As/InP single QW structure grown on a (100)-oriented *n*-type sulfur-doped InP substrate by metal organic chemical vapor deposition. The 3.5 nm undoped well layer is sandwiched in two 200 nm undoped InP layers, and on top of the structure, the 800 nm *n*-type InP cap layer is doped with Si to $2 \times 10^{18} \text{ cm}^{-3}$.

The plasma enhancement effect on QWI was investigated using this technique. The argon inductively coupled plasma (ICP) process was carried out in Plasmalab System 100 for 1 min. The rf power and ICP power were 400 and 500 W, respectively. The rapid thermal annealing (RTA) processes with a series of durations were then performed under flowing N₂ ambient. InP proximity caps were used to prevent sample surfaces from out-diffusion during annealing. Finally, samples were cleaved along the (110) surface, from which PPL spectra were measured. Three sets of experiments were carried out with the following conditions: (i) RTA only at 600 °C, (ii) ICP exposure and 600 °C RTA, and (iii) ICP exposure and 550 °C RTA, respectively.

In the measurement setup for edge-emitting PPL spectra, a 1064 nm crystal laser is used for excitation. By using a long-pass filter with a high reflection range of 400–1100 nm as a reflector, the excitation light is directed into a 40× objective lens and focused on the edge facet of the sample. The luminescence from the sample is collected by the same objective lens, and then passes the long-pass filter, a rotatable linear polarizer, and a chopper sequentially before reaching a monochromator. An InGaAs detector is used to measure the PL intensity.

By rotating the linear polarizer, the TE or TM polarization can be selected, as denoted in the inset of Fig. 1. The measured TE- and TM-PPL spectra of samples experienced ICP exposure and 600 °C RTA, as shown in Fig. 1. The annealing durations are 30, 60, 90, and 120 s. Theoretically the strengths of C-HH and C-LH transitions are

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FIG. 1. (Color online) Measured TE- and TM-PPL spectra for different annealing durations for the samples that experienced ICP exposure and RTA at 600 °C. The inset shows the schematic of pumping and notations of PL polarizations measured from an edge facet.

$$\left| \left\langle iS \uparrow | p_x | \frac{3}{2}, \frac{3}{2} \right\rangle \right|^2 = \frac{1}{2} |p_{cv}|^2 \text{ and } \left| \left\langle iS \uparrow | p_x | \frac{3}{2}, \frac{1}{2} \right\rangle \right|^2 = \frac{1}{6} |p_{cv}|^2$$

for in-plane (xy) polarization (TE) and

$$\left| \left\langle iS \uparrow | p_z | \frac{3}{2}, \frac{3}{2} \right\rangle \right|^2 = 0 \text{ and } \left| \left\langle iS \uparrow | p_z | \frac{3}{2}, \frac{1}{2} \right\rangle \right|^2 = \frac{2}{3} |p_{cv}|^2$$

for z polarization (TM), where p_{cv} is the transition matrix element between a conduction band with unit cell function $|c\rangle$ and a valence band with unit cell function $|\nu\rangle$. In our experiment, the polarization of incident excitation beam, which influences the degrees of polarization of heavy- and light-hole transitions,¹³ was not controlled intentionally. Also, the excited LHs can relax to HHs to enhance C-HH emission, whereas interface roughness may cause depolarization. Due to these factors, the observed PL spectra may not retain the relative relations of C-HH and C-LH intensities depicted by the above equations. However, it is still clearly seen that the PL spectra are highly polarization dependent. The TE-PL and TM-PL spectra reveal dominant contributions of C-HH and C-LH ground state transitions, respectively. The peak wavelengths for C-HH and C-LH ground state transitions are determined by fitting two-peak Gaussian curves to the TE- and TM-PPL spectra correlatively.

Figure 2 shows the blueshifts of the C-HH and C-LH transition wavelengths for different annealing durations for the three sets of samples. Control samples without exposure to plasma always exhibit a lower degree of intermixing than the samples treated by plasma, whereas no shifts were observed for the RTA-only samples annealed at 550 °C. In contrast to that obtained from surface emitting PL spectra, e.g., Fig. 5 in Ref. 1, this figure does not only show the trend of the blueshift, but also tells the difference between C-HH and C-LH shifts. The difference between C-HH and C-LH shifts is apparently larger for the samples that experienced ICP exposure and RTA at 600 °C than those for the other two sets of samples for short annealing time, but this diversity becomes less significant for longer time.

To further understand this, L_V and L_{III} are determined from C-HH and C-LH transition wavelengths. Figure 3

FIG. 2. (Color online) C-HH (solid lines) and C-LH (dash lines) blueshifts for different annealing durations.

shows the contour plots of C-HH and C-LH shifts with L_V and L_{III} as variables. Within the area of interest, every two contour curves have no more than one intersection, denoting that the solution of L_V and L_{III} from wavelength shifts is unique. The L_V - L_{III} loci of the three sets of samples plotted in Fig. 3 reveal the differences in time progression of QWI due to the plasma enhancement effect during RTA at 600 °C and that due to different RTA temperatures in plasma-processed samples. In the early stage of 600 °C RTA, the interdiffusion enhancement due to the ICP process is obviously much stronger on the group V sublattice. This is due to the abundance of group V defects, mostly P vacancies, in the near surface of the InP cap layer as a result of P atoms being preferentially sputtered during an ICP process.¹⁴ In the late stage after a period of annealing, however, the enhancement effects become almost equal on both sublattices, showing a quick consumption of P defects or a quick recovery of group V lattice after an annealing period.

The annealing temperature also has an effect on the situation. Even with abundance of P defects, annealing at 550 °C does not present stronger enhancement on the group

FIG. 3. (Color online) Contours of C-HH and C-LH blue shifts vs L_V and L_{III} . The L_V - L_{III} loci are derived from the experimental data for annealing durations of 30, 60, 90, and 120 s.

FIG. 4. (Color online) Calculated diffusion-length ratio vs RTA duration.

V sublattice as significantly as that at 600 °C in the early stage. This is because low temperature annealing favors group III interdiffusion due to its lower activation energy,¹⁵ and in some situation redshift may even result from group III intermixing.³

Plots in Fig. 3 show that simply presenting a k value, which is the slope of the line connecting a measured point to the origin, does not tell the whole truth about intermixing. Figure 4 shows that the calculated k is dependent on annealing time, while a near unity k is reached for long time annealing in all the three cases. k is greater than 1 in the early stage of annealing for the ICP processed samples. For RTA-only samples, k is kept nearly constant, but the initial k value is still slightly greater than 1, possibly as a result of native P vacancies in the surface atomic layers of InP (Ref. 16) for the similar reason.

In summary, diffusion lengths L_V and L_{III} are resolved from edge-emitting TE- and TM-PPL spectra for intermixing of a quaternary InGaAs/InP QW. A L_V - L_{III} locus plotted on the blueshift contour diagram clearly illustrates the time progression of a QWI process. It has been revealed that ICP exposure enhances interdiffusion on the group V sublattice more significantly in the early stage of annealing due to the

abundance of P defects in the near surface of the InP cap layer, but the reduction of P defects occurs as the annealing goes on. This results in a larger initial k but a near unity k after a period of annealing. This work presents an easy and practical approach to analyzing a QWI process for a quaternary material system.

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