STUDIES ON INTERDIFFUSION INDUCED CHANGES IN THE PHOTOLUMINESCENCE OF $In_XGa_{1-X}As$ BASED QUANTUM WELLS AND DOTS

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ABSTRACT

Interdiffusion in III-V nanostructures may occur during growth and subsequent processing steps. It is necessary to understand the process of interdiffusion in depth to control interdiffusion, which may create problems in the operation of the ultimate devices. In this paper we will try to correlate the complicated energy bands of the nanostructures with the observed optical phenomenon for explanations.

INTRODUCTION

Quantum wells (QWs), quantum wires (QWRs) and quantum dots (QDs) of III-V compound semiconductors are being progressively used in the fabrication of optoelectronic devices to increase the efficiencies and to decrease the threshold currents, taking advantage of the modified density of states of nanostructures. Interdiffusion in $In_XGa_{1-X}As$ / GaAs QWs and QDs may occur during growth and subsequent high temperature processing. Due to intermixing and redistribution of indium, strain and energy band structures of the nanostructures are liable to change after thermal cycling, leading to changes in the optoelectronic properties of the device. It is necessary to understand interdiffusion qualitatively and quantitatively to control interdiffusion, to remove the disadvantages and to use the advantages of interdiffusion favourably.

Photoluminescence (PL) has emerged as a very useful and widely used tool for studying the optical properties of III-V nanostructures [1,2,3], since a lot of information and conclusions can be determined from the PL spectra and the set up is simpler compared to transmission electron microscopy, secondary ion mass spectroscopy and Auger electron spectroscopy. Results of annealing have been reported by Sang-Wan Ryu *etal* [1] for $In_XGa_{1-X}As$/GaAs QWs of different thicknesses. On annealing, due to interdiffusion of In the PL peak undergoes a progressive blue shift and the rate of change decreases with time. There is no appreciable change of the full width at half maxima (FWHM) of the PL spectra. There are a number of reports [4-7] on the annealing of III-V quantum dots. The PL peaks undergo blue shifts and the line-widths decrease monotonically with increasing annealing time and temperature. In this paper we explain the changes in the PL spectra, of QWs and QDs on annealing through a quantum mechanical model taking into account changes of the shapes of the energy bands and levels of the energy band structures.

EXPERIMENT AND OBSERVATIONS

Annealing Of Quantum Wells

When a QW of III-V compound semiconductor like $In_X Ga_{1-X}As$ / GaAs is annealed either in a conventional electrical furnace [1] or by rapid thermal annealing (RTA) [4], In diffuses out and as depicted in Fig.1, the QW gradually widens and the band gap of the well material increases. This causes the PL peak to shift to higher energies as is shown in Fig.2. With progressive annealing, when either the time and /or temperature increases, it is observed that the rate of change of the blue shift of the PL peak decreases. The out diffusion of In is shown in Fig.3 which is calculated from the equation [1]

$$C(z) = (C_0/2)[\mathrm{erf}((h - z)/L_D) + \mathrm{erf}((h + z)/L_D)] \qquad (1)$$

where C_O is the initial In concentration, 2h the well thickness, z is the distance in the growth direction with z = 0 at the well center, and $L_D = 2\sqrt{Dt}$ is the diffusion length, where t is the annealing time and D is the diffusivity.

The observations of annealing and interdiffusion of $In_XGa_{1-X}As$/GaAs QWs as reported [1,4,8,9], agree on the point that the PL peak blue shifts and saturates, but there is a big contradiction as regards the change in

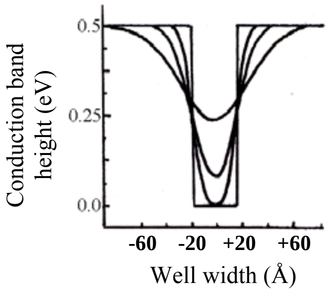

Fig.1: Gradual widening of the well width.

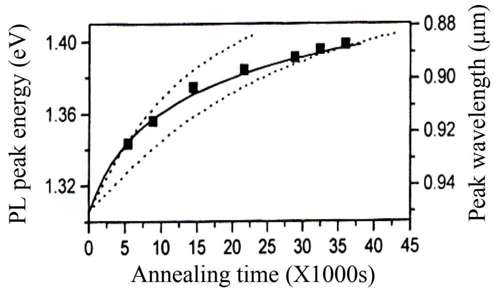

Fig.2: The dependence of the PL peak energy shift on annealing time.

the line width of the PL spectra i.e. the full width at half maxima (FWHM). According to investigations [1,4] there is no significant change of FWHM on annealing but according to other reports [8,9], the FWHM gradually broadens while the integrated PL intensity remains almost constant.

Annealing Of Quantum Dots

The PL of $In_XGa_{1-X}As/GaAs$ QDs on annealing reveals a totally different picture as compared with QWs. The initial observed broad PL spectrum which is attributed to the size distribution of QDs, after annealing undergoes a blue shift and the FWHM decreases very significantly while there is an increase in the intensity. This phenomenon continues with progressive diffusion lengths until it reaches almost saturation [4-6]. This is depicted in Fig. 4. It is also observed that the PL of QDs bigger in size start at lower energy and undergo a larger blue shift as compared with smaller dots. The observed effects of interdiffusion as reported and discussed by many research workers in the field are extremely complicated and complex. The redistribution of strain and size in QDs on annealing have been mostly thought to be responsible for the observed phenomenon [4-6] which do not seem to explain the effects very clearly.

EXPLAINATIONS FOR THE OBSERVED PHOTOLUMINESCENCE PHENOMENA

Quantum Wells

Referring to Fig. 1 and Fig. 3 we find that a given rectangular QW gradually broadens out on annealing and with progress of time the diffusion varies according to Fick's law. For strained QWs a modification has been introduced to account the initial high diffusion rate with an enhanced initial diffusion constant. Fig. 5 shows the theoretical results of the energy levels for rectangular wells of finite height and different well widths. The curves a_1 shows the variation of the sum of the energies of the first electron sub-band (e_1) and the first hole sub-band (h_1) for a QW of $In_{0.2}Ga_{0.8}As/GaAs$. a_2 shows the same results for the second levels. The band offset has been considered to be $E_C : E_V = 60 : 40$. $m_e^* = 0.059m_0$, $m_h^* = 0.44m_0$. The curves b_1 and b_2 are similar but for a QW of $In_{0.8}Ga_{0.2}As/GaAs$ with appropriate effective masses [10].

An explanation of the contradictory observations regarding the broadening of the FWHM of QWs seems to emerge from Fig. 5. The PL at any well width can be found from the band gap of the well material with a_1, b_1, a_2, and b_2 as is appropriate. It is clear from the figure that at narrow well widths only the first sub-bands exist in the well from which transitions take place. So the PL FWHM is narrow. The existence of second sub-bands only comes into the picture after the well width crosses 80 Å or more when a second set of transitions to broaden the PL FWHM is expected. So if the initial well width is small and after annealing if the well width maintains a value where it does not accommodate the second sub-band, broadening of the FWHM is not expected. On the other hand if the width of the well enters the region where the second sub-band is present,

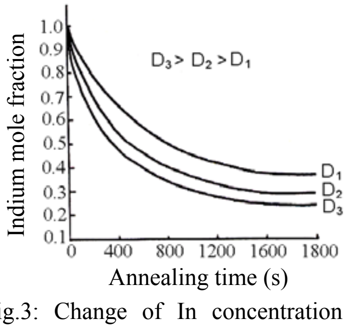

Fig.3: Change of In concentration with annealing time for different diffusivities.

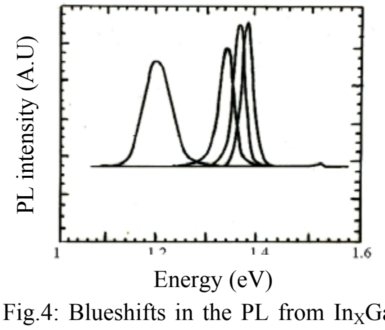

Fig.4: Blueshifts in the PL from $In_XGa_{1-X}As/GaAs$ QDs on annealing

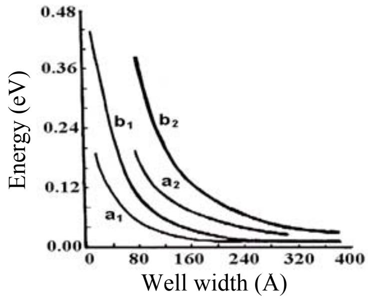

Fig.5: Variation of energy levels, (e + h). a, b for x=0.2, 0.8. 1, 2 for n=1, 2.

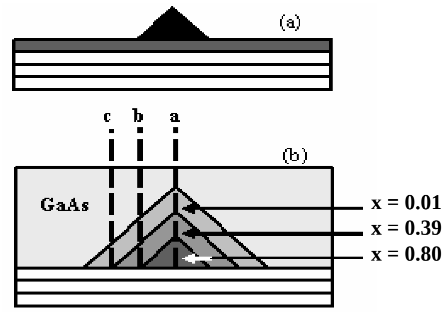

Fig.6: Growth of the Quantum Dot.

there will be a second set of close transitions which will broaden the FWHM. The exciting pump power of the PL remaining the same, the pumped up electrons will be distributed in the two levels and there are high chances that the integrated PL intensity is expected to remain more or less the same.

Quantum Dots

For $In_XGa_{1-X}As/GaAs$ QDs, on annealing the PL peak shifts towards the blue with a significant reduction in the full width at half maxima (FWHM) as shown in Fig.4. For the explanations of such observations redistribution of strain and dot size has been taken into account but a very important point has been overlooked. As outlined very clearly [11,12], when an $In_XGa_{1-X}As/GaAs$ QD emerges from a strain layer crossing the critical thickness, initially a small three dimensional pyramid highly rich in In is formed on the strained layer as shown in Fig. 6(a). As the QD grows in size the basic In rich nucleus is covered by $In_XGa_{1-X}As$ layers which progressively become depleted of In as depicted in Fig. 6(b), and the In rich bottom strained layer loses its thickness. The dot is then covered with GaAs. If we take a close look at the structure of the conduction bands of the as-grown sample at sections at various points across the QD from the substrate side, the conduction band will have the nature of an asymmetric triangular well as shown in Fig. 7. The triangular wells at different parts of the sample will vary widely in depth and width due to the variation of In and the dimension of the section under consideration. These variations will be more or less continuous. On annealing, In will diffuse from the central core to the other parts of the dot to homogenize the distribution of In throughout the dot which will ultimately lead to a rectangular well of reduced height for each triangular well as depicted in Fig.7. Model calculations were carried out for a typical pyramidal QD of square base, the height being equal to 25nm and the base being equal to 40 nm. To keep the complexity of computations within bounds, the variation of In was assumed in three phases from the central core to the outer periphery to be x=0.8, 0.4, and 0.01 respectively. After annealing the In homogenizes throughout the QD to a value of x=0.14 with no out-diffusion into the barrier and after long annealing, in the limit, the triangular wells of the conduction band are transformed to rectangular quantum wells through trapezoidal ones. It has been pointed out [13] that a trapezoidal well intermediate to the triangular and rectangular well shows intermediate PL properties. The strained band gap, E_g, of $In_XGa_{1-X}As$ is determined from the empirical relation [9]

$$E_g = 1.516 - 1.214x - 0.264x^2 \qquad (2)$$

where x is the mole fraction of indium. The energy levels of the finite triangular and the rectangular QWs of different depths and widths were calculated from the formulae, [14,15] outlined below:

$$E_n = [(V_0\hbar/a)^2 / (2m^*)]^{1/3} \times \alpha_n, \qquad (3)$$

$$E_n = \frac{2P^2}{(P+1)^2}\left[\left(\frac{n\pi}{2}\right)^2 - \frac{1}{3(P+1)^3}\left(\frac{n\pi}{2}\right)^4 - \frac{27P-8}{180(P+1)^6}\left(\frac{n\pi}{2}\right)^6\right] \qquad (4)$$

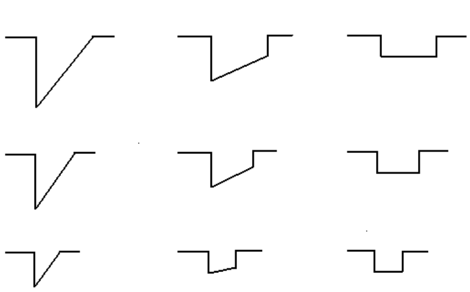

Fig.7: The conduction band changes from triangular to rectangular on annealing.

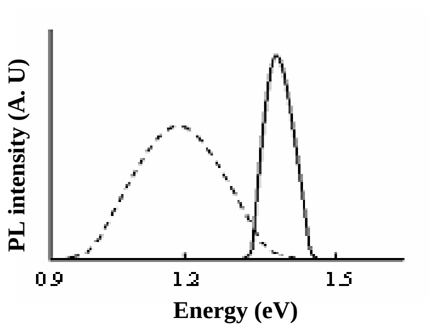

Fig.8: The results of computations of the PL energies before and after annealing.

where $\alpha_n = [3\pi(n-1/4)/2]^{3/2}$, P= $\left(\sqrt{2m^*V_0}/\hbar\right)a/2$, V_0 and a are the height and width of the well respectively and m* is the effective mass of the carrier taken into consideration. For asymmetric triangular wells, the effective masses of electrons, m^*_e, and holes, m^*_h, varied from $0.035m_0$ to $0.066m_0$ and $0.041m_0$ to $0.450m_0$ respectively, with the indium concentrations varying from 0.8 to 0.4 and in case of the rectangular wells, m^*_e and m^*_h were taken to be $0.067m_0$ and $0.45m_0$ respectively. The band offset ratio, $E_c : E_v$ was considered to be 60:40. Computations are for 6K, around which most PL experiments are carried out.

For the asymmetric quantum well representing the as grown sample the highest and lowest PL energies computed were 1.340eV and 0 .949eV.respectively. As for the rectangular wells of constant Indium mole fraction 0.41 and different widths representing the annealed QD, the highest and the lowest PL peak energies as computed were 1.440eV and 1.359eV respectively, other transitions lying in the energy interval in both the cases. It can be clearly seen that the transitions in the case of the rectangular wells take place at higher energies in a smaller energy interval to make the spectrum after annealing blue shifted, narrow and more intense. A schematic picture is depicted in Fig.8.

CONCLUSIONS

Changes of the PL spectra observed on annealing of $In_XGa_{1-X}As/GaAs$ QWs and QDs have been explained. For QWs, the PL peak shifts towards blue from increase in the bandgap due to outdiffusion from the well. The increase in the FWHM seems to arise from more than one set of transition level in the annealed, thicker QW. As for the QDs in addition to the changes in the strain distribution on annealing the triangular shaped energy bands of the as grown QDs move towards rectangular .The available transition levels decrease and bunch up in a small energy interval to decrease the linewidth with an increase in the intensity.

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