Analytic Expressions for Interdiffusion Coefficients in Quantum Well Heterostructures

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We propose simple and approximate analytic expressions to estimate the interdiffusion coefficient (D) of partially disordered quantum well heterostructures (QWHs), directly from the measurement of the photoluminescence peak shift ($\Delta h\nu$) associated with layer interdiffusion. QWHs of two III-V compound semiconductor systems were investigated: $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ ($x \approx 0.2$) and $\mathrm{GaAs/Ga}_{1-x}\mathrm{Al}_x\mathrm{As}$ ($x \approx 0.3$). By assuming the envelope function approximation and Fick's law, we have calculated $\Delta h\nu$ as a function of the interdiffusion length $L_D = (Dt)^{1/2}$, where t is the interdiffusion time, considering QW thiknesses (L_z) in the range of previous litterature data. A simple relationship was obtained for the variation of $\Delta h\nu$ with the dimensionless parameter L_D/L_z in each system, thus providing simple expressions for D as a function of $\Delta h\nu$, L_z and t. Within a factor of two, these expressions satisfactorily account for most D values previously reported, in the range of as-grown compositions and L_z values considered for each system.

I. Introduction

Considerable attention has been given in recent years to the investigation of partial layer interdiffusion in III-V compound semiconductor quantum well heterostructures (QWHs)[1-6]. This phenomenon has found important applications in the fabrication of optoelectronic devices because high temperature anneals are often required during processing, such as epitaxial overgrowth and ohmic contact alloying. In addition, the QW shape can be intentionally altered by apropriate thermal treatments, thus enabling the fabrication of QW lasers with adjusted emission wavelength[1,2]. The layer interdiffusion process is known to change the optical properties of QWHs, resulting in a shift of the photoluminescence (PL) peak associated with the ground state emission to higher energies. The interdiffusion coefficient (D) is generally determined by solving Schrödinger's equation for the QWH, assuming Fick's law for the intermixing process, and adjusting the PL data with the interdiffusion length defined as $L_D = (Dt)^{1/2}$, where t is the interdiffusion time. This procedure accounts for a wide range of D values obtained in QWHs for various III-V compound semiconductor systems[3-6], however it is time consuming, somewhat complicated and unpractical for a rapid and approximate analysis.

In the present work, we propose simple and approximate expressions to determine the interdiffusion coefficient of partially disordered QWHs from the direct measurement of the shift of the PL peak ($\Delta h\nu$) to higher energies associated with the interdiffusion process. Two important III-V compound semiconductors were analysed in the composition range and QW thicknesses of interest for optoelectronic device applications: the strained layer $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ ($x \approx 0.2$) system and the lattice matched $\mathrm{GaAs/Ga}_{1-x}\mathrm{Al}_x\mathrm{As}$ ($x \approx 0.3$) system. We have calculated from first principles $\Delta h\nu$ as a function of L_D considering values of the as-grown QW thickness (L_z) and $\Delta h\nu$ in the range where most previous data has been reported. Simple expressions were obtained for the variation of $\Delta h\nu$ with the dimensionless diffusion parameter L_D/L_z in each system, thus providing a direct relationship for D as a function of $\Delta h\nu$, L_z and t. These expressions satisfactorily account for previously reported data where partial interdiffusion may be useful for device applications.

II. Theoretical analysis

The interdiffusion process in QWHs may be regarded as the transition of an initially as-grown abrupt rectangular compositional profile to a final graded compositional profile. The QW emission is the sum of the first electron subband energy, the first heavy hole subband energy and the band gap of the material inside the well. We have calculated the ground state emission in QWHs presenting an abrupt rectangular compositional profile, as well as in disordered graded compositional profiles. First, we shall describe QWHs in the strained layer $In_xGa_{1-x}As/GaAs$ system. Since the $In_xGa_{1-x}As$ QW layer is under a compressive strain relative to GaAs, the hydrostatic component of the strain increases the fundamental band gap with respect to the unstrained material, while the biaxial compression lifts the degeneracy of the heavy and ligth hole valence bands. The shift of the heavy hole valence band is smaller than that of the light hole valence band, so the ground state emission involves only the first heavy hole subband. The expressions used for the compositional dependence of the band gap of the unstrained material, the band gap change due to the strain, and the interpolating parameters of GaAs and InAs are those reported in ref.[7]. The band gap was adjusted with the strain, and we have assumed a 70/30 split for the conduction/valence band offset[7]. The ground states of the QWHs both abrupt and graded were calculated in the framework of the effective mass theory using the envelope function approximation[8]. In the final graded profile we have assumed Fick's second law of diffusion and the fact that In and Ga have the same diffusion coefficient, which is isotropic and independent of x. The final graded profile is represented by the superposition of two complementary error functions[4,7,9]:

$$x(z) = (x_0/2)\{erf[(\{L_z/2\} - z)/2L_D] + erf[(\{L_z/2\} + z)/2L_D]\}\ , \qquad (1)$$

where z is the growth axis and x_0 is the as-grown composition of the QW material. In the compositionally graded profiles, we have undertaken a standard numerical calculation of the electron and heavy hole ground state energies considering both the strain and quantum size effects[7]. In the calculations we have also included the changes of the effective mass and strain as a function of composition in the barrier layers. We have also calculated under the same assumptions the PL peak shift to higher energies induced by layer interdiffusion in QWHs of the $GaAs/Ga_{1-x}Al_xAs$ system. A 60/40 split for the conduction/valence band offset was used in the calculations with the material parameters reported in ref.[10]. In the calculations of the disordered QWHs we have also included the effective mass change as a function of the composition of the barrier layers. In this case the initial composition x_0 originates from the barrier layers, and therefore the final graded profile is represented by a slightly different expression, which is given by[3,11-13]:

$$x(z) = x_0\{1 - (1/2)[erf\{[(L_z/2) - z]/2L_D\} + erf\{[(Lz/2) + z]/2L_D\}]\}\ . \qquad (2)$$

III. Results

III.1. $In_xGa_{1-x}As/GaAs$ QWHs

The results of the calculated PL peak energy shifts $\Delta h\nu$ to higher energies as a function of L_D for $x_0 = 0.20$ and $L_z = 100$ Å are presented in figure 1. The dependence of the confinement energies of the electron and heavy hole in terms of L_D are also shown. After the layer interdiffusion process, the energy separation of the electron and heavy hole subband increases, resulting in a shift of the ground state emission to higher energies. One notices that the confinement energies of the electron and heavy hole increase with the diffusion

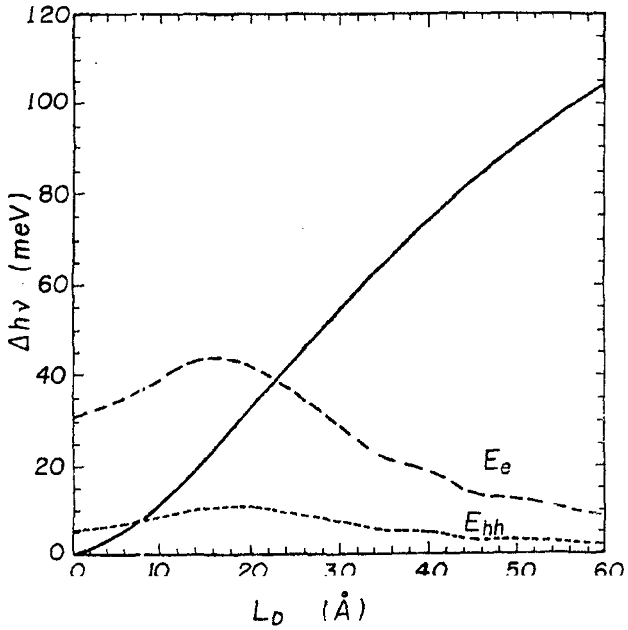

Figure 1: Plot of $\Delta h\nu$ versus L_D for an $In_xGa_{1-x}As$ / GaAs ($x = 20$) QWH with $L_z = 100$Å. Also shown are the confinement energies of the electron (E_e) and the heavy hole (E_{hh}).

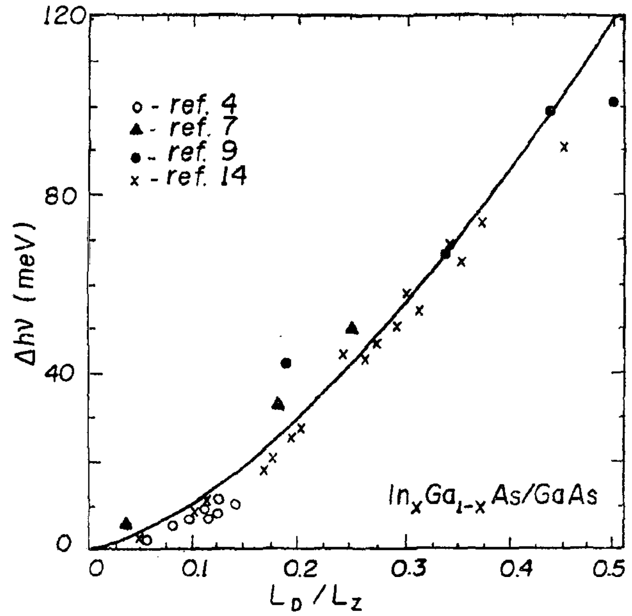

Figure 2: Plot of $\Delta h\nu$ versus L_D/L_z for an $In_xGa_{1-x}As$ / GaAs ($x = 20$) QWH (solid line). Data points are from: ref.[4] (open circles) with $x_0 = 0.20 - 0.24$ and $L_z = 60 - 100$Å. ref.[7] (closed triangles) with $x_0 = 0.23 - 0.24$ and $L_z = 80 - 100$Å, ref.[9] (closed circles) with $x_0 = 0.23$ and $L_z = 80$Å, and $L_z = 80$ Å, and ref.[14] (crosses) with $x_0 = 0.20$ and $L_z = 100$Å.

length until approximately one third of the well thickness. At these diffusion lengths the peak shifts due to an increase in confinement energy of the particles and to a decrease in In concentration at the centre of the well are approximately equal. The main contribution to the peak shift for the smaller interdiffusion lengths is the change in confinement energy, while for larger interdiffusion lengths is an outdiffusion of In from the QW layer.

When L_z decreases, $\Delta h\nu$ increases more steeply as function of L_D (data not shown), because a smaller QW exhibits a larger increase with L_D than a larger QW. However, for very large values of L_D there is a transition after which the opposite behaviour is obtained. The as-grown QW transition energy increases with a decrease in L_z, and because the QW ground state energy must be always below the bandgap of the barrier layers, $\Delta h\nu$ saturates to a lower value for a smaller L_z. Since we are mainly interested in the range of low $\Delta v\nu$ and L_D values, this point will not be discussed further.

In order to compare data with different QW thicknesses, we have evaluated the PL peak energy shift as a function of the dimensionless diffusion parameter L_D/L_z. Figure 2 presents the calculated curve of $\Delta h\nu$ in terms of L_D/Lz. In this case! a common behaviour is obtained for the variation of $\Delta h\nu$ with different values of L_z in the range 60-100Å with $x_0 = 0.20 - 0.24$, which represents reasonably well the previous data reported by various investigators. These data correspond to measured peak shifts with calculated values of L_D of single QWHs[7,9,14] as well as superlattices[4]. In references where L_D data were not available, they were obtained from the relation $L_D = (Dt)^{1/2}$. The change in x_0 affects primarily the barrier height, which induces a larger increase of $\Delta h\nu$ with L_D going from 0.2 to 0.24, but the net effect has a minor contribution compared to the spread of the reported data. Although $\Delta h\nu$ saturates at higher values, the common trend obtained in terms of L_D/L_z in the range shown in figure 2 may be approximated by a simple exponential expression having the following form:

$$\Delta h\nu \approx k(L_D/L_z)^{\beta} , \qquad (3)$$

where $k = 345$meV and $\beta = 1.52$ if $\Delta h\nu$ is expressed in meV. These values of k and β are valid only in the range of x_0 and L_z values shown in figure 2. k and β are primarily dependent on the barrier height of the QWH, and consequently increase with x_0. In fact, lower values of these fitting parameters are obtained for $x_0 = 0.14$, where $k \approx 160$meV and $\beta \approx 1.27$. k and β also present a small increase with L, in the range of values shown in figure 2, which can be neglected on a first approximation analysis. Hence, a direct relationship can be extracted for D as a function of $\Delta h\nu$, L_z and t, for the data shown in figure 2:

$$D \approx (L_z^2/t) \times (\Delta h\nu/k)^{2/\beta} \ . \tag{4}$$

The In-Ga interdiffusion coefficients in $In_x Ga_{1-x}As$ / GaAs QWHs can therefore be roughly calculated with this simple relation from the knowledge of the as-grown QW thickness L_z, the interdiffusion time t, and the measurement of $\Delta h\nu$. Most of the D data previously reported shown in figure 2 are less than a factor of two from the calculated values using the simple formula shown above.

III.2 - GaAs/$Ga_{1-x}Al_xAs$ QWHs

We have obtained a similar behaviour for $\Delta h\nu$ as a function of L_D shown in figure 1 (data not shown) for QWHs of the GaAs/$Ga_{1-x}Al_xAs$ system, but with a larger increase of the peak shift in terms of L_D. This larger shift occurs because the GaAs/$Ga_{1-x}Al_xAs$ system is lattice matched, and therefore the band gap change due to strain which reduces the PL peak shift in a compressively strained QWH is absent in this case. Figure 3 presents the calculated curves of $\Delta h\nu$ as a function of the dimensionless parameter L_D/L_z for GaAs/$Ga_{1-x}Al_xAs$ QWHs for two values of the barrier layers composition: $x_0 = 0.25$ and $x_0 = 0.3$. Also shown are previous data reported by various investigators corresponding to measured peak shifts with calculated L_D values, where most data are with $x_0 = 0.3$ and L_z in the range 80-160Å [1,3,12,13], but there are also some data with lower L_z [3] and with $x_0 = 0.25$ and L_z = 130Å [11]. The data can be satisfactorily represented by the same expression of $\Delta h\nu$ in terms of L_D/L_z given above for

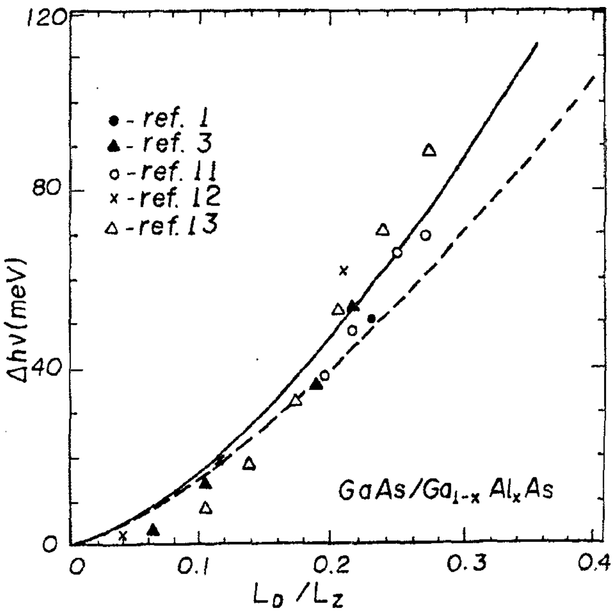

Figure 3: Plot of $\Delta h\nu$ versus L_D/L_z for a GaAs / $Ga_{1-x}Al_xAs$ QWH with $x = 0.30$ (solid line) and $x_0 = 0.25$ (dashed line). Data points are from: ref.[1] (closed circle) with $x_0 = 0.30$ and L_z = 85Å, ref.[3] (closed triangles) with $x_0 = 0.30$ and L_z = 54 − 160Å, ref.[11] (open circles) with $x_0 = 0.25$ and L_z = 130Å, ref.[12] (crosses) with $x_0 = 0.32$ and L_z = 80Å, and ref.[13] (open triangles) with $x_0 = 0.30$ and L_z = 150Å.

strained QWHs, but with different fitting parameters k and β. In figure 3, the solid line ($x_0 = 0.3$) can be best fitted with $k \approx 540$meV and $\beta \approx 1.5$, and the dashed line ($x_0 = 0.25$) with $k \approx 390$meV and $\beta \approx 1.4$, if $\Delta h\nu$ is expressed in meV. However, a better fit of the data, previously reported in the range of low $\Delta h\nu$ may be obtained with $k \approx 720$meV and $\beta \approx 1.75$. The same considerations discussed above apply here to k and β. The change in QW width has a minor effect compared to the change of barrier height and also with the spread of the data points shown in figure 3. The Al-Ga interdiffusion coefficients can therefore be approximately calculated from the simple relation above with the knowledge of $\Delta h\nu$, L_z and t, in the range of x_0 and L_z considered in figure 3. Most of the D values thus obtained with the appropriate fitting parameters k and β are within a factor of two from the true data points.

IV. Conclusion

In conclusion, we propose simple and approximate expressions to determine D from the measurement of

$\Delta h\nu$ induced by layer interdiffusion. QWHs of the strained layer $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ ($x \approx 0.2$) system and of the lattice matched $\mathrm{GaAs/Ga1} - x\mathrm{Al}_x\mathrm{As}$ ($x \approx 0.3$) system were investigated. We have calculated $\Delta h\nu$ for given values of L, and L_D, and we have obtained for each system, a simple expression for the variation of $\Delta h\nu$ with the dimensionless diffusion parameter L_D/L_z. These expressions provide a direct relationship for D in terms of $\Delta h\nu$, L, and t, which satisfactorily account for most previous reported data in the range of as-grown compositions and L_z values considered in this study. Finally, these simple formulas may be extended to a wider range of $\Delta h\nu$ and L_D values, as well as to other III-V compound QWH systems, by an apropriate modification of the fitting parameters k and β which can be obtained from similar calculations described here.

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