Type II narrow double barrier quantum well structures: Γ -X coupling and interface effects

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Abstract. - Photoluminescence (PL), PL excitation and time resolved PL experiments have been performed on $\text{Al}_{0.42}\text{Ga}_{0.58}\text{As}/\text{AlAs}/\text{GaAs}$ symmetric double barrier quantum wells (DBQW) with only one or two AlAs monolayers constituting the intermediate barriers. In agreement with the envelope function predictions we show that such DBQW's undergo a type I - type II transition when the GaAs thickness is reduced below 7 and 5 monolayers for 2 and 1 AlAs molecular planes respectively. In type II configuration the PL decay time is found to be strongly dependent on the energy difference between AlAs X_z - and GaAs Γ - electron confined states and the coupling parameter of the Γ and X_z valleys can be deduced (4.2 meV on average). Similar experiments on DBQW's grown on 2° off (001) vicinal surfaces show that the exciton lifetime remains close to that of nominal samples. Considering also former results obtained in type II GaAs/AlAs superlattices, it is concluded that the Γ -X mixing potential in quantum structures is intrinsically linked to the AlAs/GaAs interface.

1. Introduction.

Mixing of the different conduction band states across heterostructure interfaces may introduce profound modifications to electronic wave functions, especially when the band energies are close together [1-2]. In the GaAs/AlAs confining systems where, by the choice of layer thicknesses, the GaAs Γ - electron states can be brought in the vicinity of AlAs X - electron states, the coupling effect has noticeable consequences on tunnelling processes[3], carrier transfer rates[4] and recombination dynamics[5-7]. Basically, the intervalley mixing is due to the perturbation produced by the potential discontinuity at the heterointerface and appears as an intrinsic phenomenon. However, interface roughness[6] or additional periodicity introduced in superlattices[8] are anticipated to have an influence on the Γ -X coupling strength. In fact, the connection between the magnitude of the potential responsible for the mixing and the interface structure have never been experimentally confirmed. This is the issue we are addressing in this paper via a comparison between the recombination properties of type II quantum wells grown on nominal and vicinal (001) surfaces. The misorientation is a means to produce a controlled interface roughness, the misorientation angle determining an average width of terraces.

2. Samples and experimental technique

The quantum structures used in this work are isolated and symmetric double barrier quantum wells (DBQW) $\text{Al}_{0.42}\text{Ga}_{0.58}\text{As}/\text{AlAs}/\text{GaAs}$ in which the electrons are confined within GaAs or in only one or two AlAs monolayers [9]. In the following, DBQW's will be designated using the notation (a-b), where a

and b are respectively the AlAs and GaAs thickness in monolayer (ML) length units. Two identical (a-b) DBQW structures were simultaneously grown by molecular beam epitaxy : one on an (001) exactly oriented substrate and the other on a substrate misoriented 2° towards the (111)A plane. The growth parameters were carefully adjusted in order to achieve a precise control of the size of wells and barriers. Details on the growth procedure was previously reported in reference [10]. CW selective and excitation photoluminescence (SPL and PLE respectively) experiments were performed with a tuneable dye laser and a 1m focal length double monochromator. For time resolved experiments, excitation light was supplied by a synchronously pumped mode-locked dye laser with a time duration of pulses of about 10 ps with a repetition rate of 76 MHz which could be reduced up to 1MHz by a cavity dumper placed after the dye laser. PL was analysed by a time correlated single photon counting apparatus providing a time resolution of 100 ps. The excitation energy was chosen just above the direct-band gap of (Al,Ga)As.

3. Band alignment in DBQWs.

The isolated DBQWs give various electronic configurations : it is possible to produce direct as well as indirect quantum structures by selecting the GaAs quantum well widths (L_w), the AlAs intermediate barrier thicknesses and Al composition (x_{Al}) of the external (Al,Ga)As barrier. In type-II DBQWs electrons can be either confined in the AlAs layers (type-II β) or delocalized in (Al,Ga)As (type-II α). In fact, the x_{Al} and L_w values for which the electron ground states are confined in AlAs (type-II β) are found in a limited range. The calculation in the envelope function approximation of the different band configurations is summarised on the Fig. 1a and 1b for QW's bordered by 1 and 2 AlAs ML's respectively.

Fig. 1 - Diagram of isolated $Al_xGa_{1-x}As/AlAs/GaAs$ DBQW electronic structure as a function of the Al content of the external barrier and the GaAs layer width (L_w). The AlAs layer thickness is a) one and b) two monolayers. The full line boundaries fix the limits of the (x_{Al}, L_w) extent where the electrons are confined in GaAs, in AlAs (type II β) or delocalized in (Al,Ga)As (type II α). hh_1 and e_1^Γ full and dotted lines indicate respectively the confinement threshold of holes and Γ electrons in the GaAs quantum well. The area in which Xz confined electron states exist in AlAs spreads between the (1) and (3) curves. For clarity the diagram relative to the Xxy conduction bands of (Al,Ga)As and AlAs are not included.

Fig. 2 - Transition energies measured at 2K by PL (triangles) and PLE (circles for $e_1^\Gamma-hh_1$ and squares for $e_1^\Gamma-lh_1$) in DBQWs as a function of the GaAs quantum well width. a) one and b) two AlAs monolayers are inserted at the GaAs/(Al,Ga)As interface. Dotted and short dashed lines represent respectively the calculated $e_1^\Gamma-hh_1$ and $e_1^\Gamma-lh_1$ direct transition energies, full and large dashed lines the indirect e^{Xz} and e^{Xxy} transitions respectively. The exciton binding energies are not included in the calculations. The splitting shown for the $e^{Xz}-hh_1$ transition on Fig. 3b recall that for narrower GaAs quantum wells the coupling between the X electron states in the AlAs layers becomes effective.

Fig. 3 - Low temperature photoluminescence decay curves of double barrier quantum wells having two AlAs monolayers inserted at the GaAs/Al_{0.42}Ga_{0.58}As interface.

Fig. 4 - Evolution as a function of the temperature of the photoluminescence decay times of (2-5) DBQW's grown on nominal (2-5)n and vicinal (2-5)v surfaces.

It can be seen that, for the optimum values of x_{Al} near 0.4, only the (1-3), (1-4), (2-4), (2-5) and (2-6) type-II β DBQW's are available.

Figure 2 compares the direct optical transitions deduced from PLE with the theoretical predictions. According to Fig. 1, the Γ ground electron states of the (1-3) and (2-4) DBQW's are found not to be confined; the e_1^Γ -hh₁ transitions appear below the barrier absorption threshold on the (1-4) and (2-5) DBQW's spectra and both e_1^Γ -hh₁, and e_1^Γ -lh₁ transitions are detected for wider GaAs QW's. In addition, no luminescence was observed for (1-2) DBQW structure in which the holes are not expected to be confined. Considering that for such narrow quantum structures the binding energies of direct excitons are nearly the same as in the barrier (around 6 meV), the comparison between the calculated band gap and the direct transition energies deduced from PLE spectra are in a quite good agreement [9]. Turning now to the type-I DBQW PL results, it should be noted that the emission lines are significantly Stokes shifted with respect to the e_1^Γ -hh₁ free exciton transitions and this is probably also the case for type-II DBQW's[11]. The low temperature PL bands of the indirect structures are clearly composed of two well separated recombination lines with an intensity ratio which depends on the injection level and temperature. These features lead to conclusion that the type-II DBQW's PL is mainly due to recombination of localised excitons and to impurity related transitions.

Figure 3 displays the time dependence at 4K of DBQW's PL. It clearly shows that the (2-6) and (2-5) DBQW's recombine on a ns timescale, as expected for type-II quantum structures, whereas the (2-7) one behaves as a direct band gap structure (on a ps timescale). In addition, the time decays τ_1 of the indirect transitions are an increasing function of the energy spacing ($\Delta_{\Gamma-X}$) between e_1^Γ (GaAs) and e_1^{Xz} (AlAs) confined electron states : the recombination time in the (2-6) DBQW where the Γ and X conduction-bands are almost aligned ($\Delta_{\Gamma-X} = 11$ meV) is shorter than in the (2-5) DBQW where $\Delta_{\Gamma-X}$ is evaluated to be 56meV. These trends manifest the influence of the conduction band valley mixing across the DBQW interfaces, i.e. the interface induced coupling between the AlAs X- and GaAs Γ -confined states. The time decay measurements performed on DBQW's grown on vicinal surfaces have shown that the interface roughness generally leads to a weak reduction of τ_1 . This variation remains stable when the temperature is raised (see Fig. 4) showing that the competition between the radiative and nonradiative lifetimes is not greatly modified by the substrate orientation. Actually, table 1 shows that the modification of τ_1 is fully accounted for by the additional lateral confinement energy in AlAs due to the step array in type II DBQW's [10].

4. Coupling potential.

According to [6], the pseudo-direct radiative lifetime τ_r is proportional to the direct radiative recombination lifetime τ_d and the squared ratio between $\Delta_{\Gamma-X}$ and the spatial overlap of electron wave functions in AlAs and GaAs scaled by the investigated coupling constant A. The overlap integral can be evaluated in the framework of the envelope function which was shown to be suitable for the prediction of the confinement energies of nominal DBQW's. On the other hand, the temperature dependence of the ratio between PL decay time τ_l and the integrated PL intensity shows that the non radiative phenomena remain negligible in all DBQW's at 4K and allow the relation $\tau_r = \tau_l$ to be used [12]. The time constant τ_d is assumed to be 250ps on the basis of time decay measurements on direct DBQW's reported in [13]. The deduced A constants are given in table 1. It should be noted that the lateral confinement due to step arrays in DBQW's grown on vicinal surfaces implies that both confinement energy and wave function are modified, even if the overlap integral does not change appreciably.

Table 1 - Γ -X Coupling parameters in Type II DBQW's deduced from PL time decay and PLE measurements. $\Delta_{\Gamma-X}$ and $\langle \Psi^X / \Psi^\Gamma \rangle$ are respectively the energy splitting between the GaAs- Γ - and AlAs-X confined ground states and the calculated overlap integral of associated wave functions.

DBQW's	$\Delta_{\Gamma-X}$ (meV)	$\langle \Psi^X / \Psi^\Gamma \rangle$	τ_l (ns) experimental	A (meV)
(2-5) n	56	0.76	70	4.4
(2-6)	11	0.81	2.7	4
(1-4)	14	0.97	2.8	4.2
(2-5) v	50	0.76	54	4.5

5. Conclusion

We have shown that the Γ -X coupling parameters in single DBQW's are nearly insensitive to layer thicknesses, interface roughness or impurities. In addition, they are very close to those measured in short period superlattices [7]. We conclude that the coupling potential in quantum structures is intrinsically linked to the AlAs/GaAs interface.

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