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Photoreflectance spectroscopy of semiconductor device active regions: quantum wells and quantum dots

GRZEGORZ SĘK, JAN MISIEWICZ

Institute of Physics, Wrocław University of Technology, Wybrzeże Wyspiańskiego 27, 50–370 Wrocław, Poland.

ALFRED FORCHEL

Institute of Physics, Würzburg University, Am Hubland, D-97074 Würzburg, Germany.

Photoreflectance spectroscopy is presented as a powerful tool of the investigation of different kinds of low dimensional semiconductor structures being a fundamental part of modern electronic or optoelectronic devices. The derivative nature of the modulation spectra gives high sensitivity of the method for optical transitions including those with nominally weak oscillator strength, like indirect in the real space or parity forbidden ones, and allows to investigate small volume objects like quantum dots. This short review includes results concerning single quantum wells of new material system like InGaAsN/GaAs, coupled quantum wells of two different material systems (GaAs/AlGaAs and InGaAs/GaAs) and several InGaAs/GaAs quantum dot structures.

1. Introduction

Investigations of low dimensional semiconductor structures have gained a considerable interest during the last decade. The interest in these structures, especially quantum wells and quantum dots, is due to their importance in understanding the fundamental processes in quantum structures and arises in part from the expectation that their electronic properties might be utilized in electronic and optoelectronic devices, *e.g.*, light emitters and detectors or very fast field effect and bipolar transistors. In the last few years coupled quantum dot structures have become the subject of special interest because of new quantum computational concepts and the possibility of implementing the structures in the construction of quantum bits [1].

In this work we present the results of photoreflectance (PR) spectroscopy applied to several kinds of low dimensional quantum well and quantum dot structures. We focus on the investigation of optical transitions. Photoreflectance as a contactless modulation technique seems to be one of the most powerful optical methods for this

purpose due to its extraordinary sensitivity to optical transitions (including those between excited states) at room temperature and even at elevated temperatures, *i.e.*, at the conditions of work of semiconductor devices.

2. Experimental

The samples investigated in this work were grown by MBE on semi-insulating (001) GaAs substrates. They are as follow:

– 9 nm thick $In_{0.28}Ga_{0.72}As_{1-x}N_x$/GaAs single quantum wells (SQW).

– $Al_xGa_{1-x}As$/GaAs double quantum wells (DQW), 7.5 nm thick each, separated by 0, 1, 3 or 5 ML of AlAs barriers.

– Coupled $In_xGa_{1-x}As$/GaAs quantum wells consisting of 7.5 nm thick $In_{0.045}Ga_{0.955}As$ wells, separated by different thickness GaAs barriers.

– $In_{0.6}Ga_{0.4}As$/GaAs quantum dot laser structure for 980 nm.

– Double quantum dot (DQD) structures consisting of two layers of self-assembled $In_{0.6}Ga_{0.4}As$/GaAs quantum dots separated by 3, 5 or 10 nm.

Details of the photoreflectance setup are described in the literature [2]–[4]. As a probe beam served a light from a halogen lamp dispersed through a double grating monochromator. The 632.8 nm line of a He-Ne laser was used as a pump beam. The reflected light was detected by a silicon or germanium photodiode.

3. Results and discussion

3.1. $In_{0.28}Ga_{0.72}As_{1-x}N_x$/GaAs single quantum well

Figure 1 shows PR spectra of $In_{0.28}Ga_{0.72}As_{1-x}N_x$/GaAs SQW samples for various nitrogen contents [5]. All the spectra are dominated by GaAs band gap bulk-like signal above the energy of 1.4 eV. Below this energy the quantum well related transitions are observed. It is seen that with the increase of the nitrogen content all the QW features shift towards lower energies. The red shift is significant because the introduction of even small amounts of nitrogen in InGaAs greatly reduces the band gap energy. In our case it is as much as 168 meV (180 nm) per 1% of nitrogen for the ground state transition. Since the well width and the indium content are unchanged, the band gap decrease with increasing N content indicates a large bowing parameter due to N incorporation.

The quantum well PR signal intensity decreases and the energetic broadening increases with increasing N concentration. Since there is a large atomic size difference between N and As, the large local strain from N incorporation into InGaAs would result in inferior crystallinity. Therefore, the InGaAsN material may have a large immiscible composition domain forming compositional clustering. This causes the alloy disorder, which can play an important role in the broadening due to the strong variation of the InGaAsN band gap with nitrogen mole fraction. In general, the more

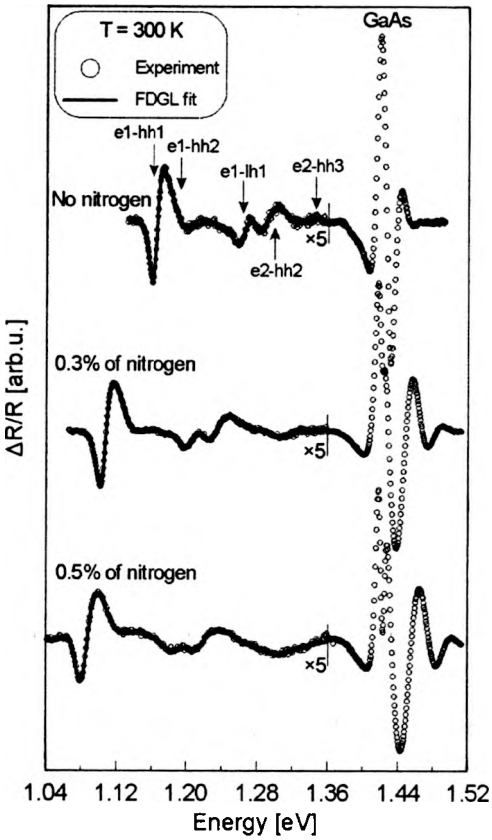

Fig. 1. Photoreflectance spectra of $In_{0.28}Ga_{0.72}As_{1-x}N_x/GaAs$ SQW samples for $x = 0$, 0.003 and 0.005 [5].

N atoms are incorporated into InGaAs, the worse is the sample quality and the larger PR broadening parameter. For example, for the fundamental e1–hh1 transition the broadening changes from 5.6 meV for the $In_{0.28}Ga_{0.72}As/GaAs$ sample, to 8 meV for the sample with 0.5% of nitrogen, and for the lowest light hole transition (e1–lh1) it increases from 11.2 to 27.0 meV for the same samples.

The experimental energies have been compared with the results of the envelope function calculations for a finite rectangular quantum well. The comparison of the theoretical dependences of the optical transition energies of the InGaAsN/GaAs SQW *vs.* N concentration with the experimental values is presented in Fig. 2 [5]. The experimental points match to calculated curves very well and the agreement is very good in spite of some approximations made in the description of InGaAsN material properties. The use of the modulation technique has allowed us to investigate, in addition to common parity allowed transitions, also forbidden transitions like e1–hh2 and e2–hh3. The observation of the latter ones has been connected with the influence

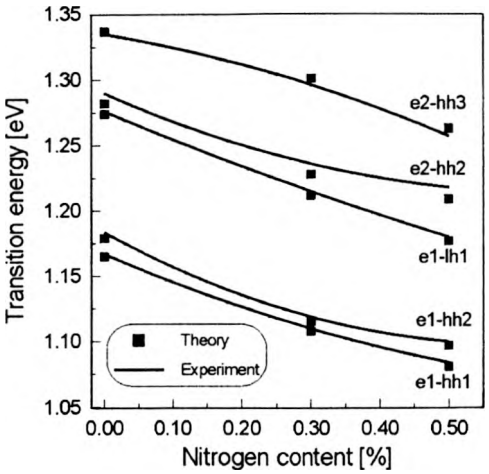

Fig. 2. Comparison of the theoretical values (solid lines) with the experimental ones (squares) of the transition energies *vs.* nitrogen content for $In_{0.28}Ga_{0.72}As_{1-x}N_x$/GaAs SQW for x = 0, 0.003 and 0.005 [5].

of the surface electric field and/or random alloy fluctuations, which could destroy the symmetry of quantum well.

3.2. $Al_{0.3}Ga_{0.7}As$/GaAs double quantum wells with AlAs separating barrier

In Figure 3, the room temperature PR spectra of GaAs/$Al_{0.3}Ga_{0.7}As$ double quantum well structures are shown for various thicknesses of AlAs separating barrier [6]. One can see strong GaAs band gap related feature and several confined state DQW transitions. Almost all allowed (between electron and hole symmetric, and antisymmetric states, respectively) transitions are observed. All the transitions are identified basing on the results of simple envelope function calculations neglecting excitonic effects, in a first approximation. The comparison of experimental and theoretical transition energy dependences on AlAs barrier thickness is presented in Fig. 4 [6]. Generally, a very good agreement has been obtained except the high energy transitions, where some discrepancy between experiment and calculation appears. This disagreement may be related to non-parabolicity effects which are more important for higher energy subbands. It can be also due to some problems with material parameters, *e.g.*, band offset ratio which was assumed to be 40% for valence band. Because there is still no full understanding of this parameter, its value in the literature can differ slightly. It influences mainly the high energy subbands located close to the edge of the potential well.

Another problem is the thickness of the separating barrier which was taken to be exactly equal to the value determined from growth conditions and some post-growth characterisation. Even a small change in the average barrier thickness influences the absolute energies of the subbands as well as the splitting between symmetric and antisymmetric states. These shifts are larger for higher index levels.

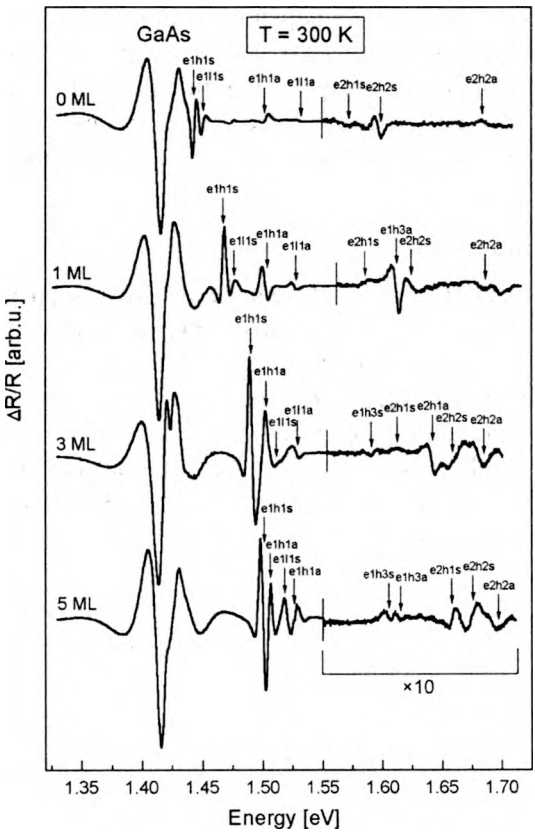

Fig. 3. Room temperature PR spectra of $Al_xGa_{1-x}As/GaAs$ DQW structures for various AlAs separating layer thickness; s – transitions between symmetric states, a – transitions between antisymmetric states [6].

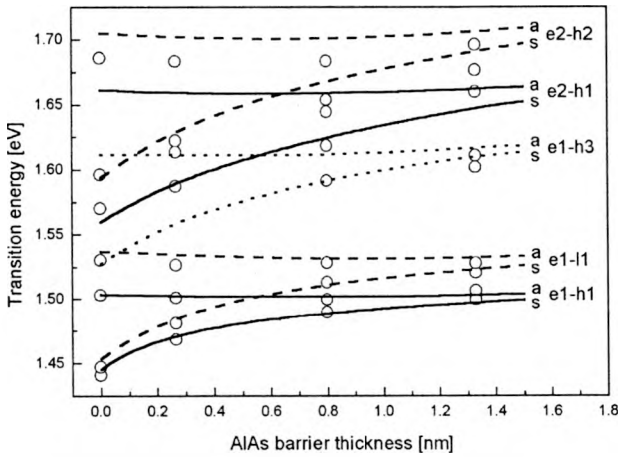

Fig. 4. Dependence of the transition energies on the AlAs barrier thickness; circles – experimental data, lines – calculation [6].

It is worth to note, what is clearly seen in Fig. 4, that depending on the thickness of the separating barrier we can have different succession of transitions, especially for the value of about 6 Å, corresponding almost exactly to the thickness of two monolayers of AlAs, where the crossings of some of the calculated curves are observed. Similar feature is also seen at about 1 Å, but this unrealistic case (below 1 monolayer thickness) is not interesting.

3.3. $In_{0.045}Ga_{0.955}As/GaAs$ double quantum well

Our calculations for $In_{0.045}Ga_{0.955}As/GaAs$ DQW show that we have only two electron and two heavy hole confined states (one symmetric and one antisymmetric), and light holes unconfined in the barriers (type II system for light holes). Therefore, we can expect at most six optical transitions in the PR spectrum: four heavy hole-related (e1s–h1s, e1s–h1a, e1a–h1s, e1a–h1a), including the parity-forbidden ones, and perhaps two indirect in the real space light hole transitions (e1s–l, e1a–l). The calculated splitting between states is large enough to neglect the mixing of symmetric and antisymmetric states, in a first approximation.

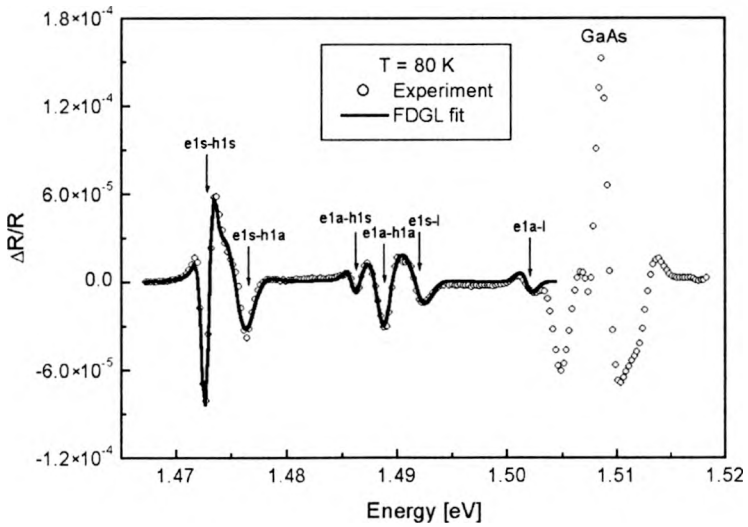

Fig. 5. PR spectrum of $In_{0.045}Ga_{0.955}As/GaAs$ DQW structure [7].

Figure 5 shows PR spectrum of the $In_{0.045}Ga_{0.955}As/GaAs$ DQW sample at 80 K [7]. Indeed, six features are observed below the GaAs bulk-like related signal. The most interesting is the region of the forbidden transitions, *i.e.*, transitions between the states of different symmetry (e1s–h1a and e1a–h1s). This kind of transition can occur because of some asymmetry of the DQW structure, like random alloy fluctuations or internal electric field, and/or mixing effects of heavy and light hole excitons. Although the DQW is buried at about 200 nm from the surface, it can still be in a weak electric field due to the band bending at the surface. It has been already shown by us that this

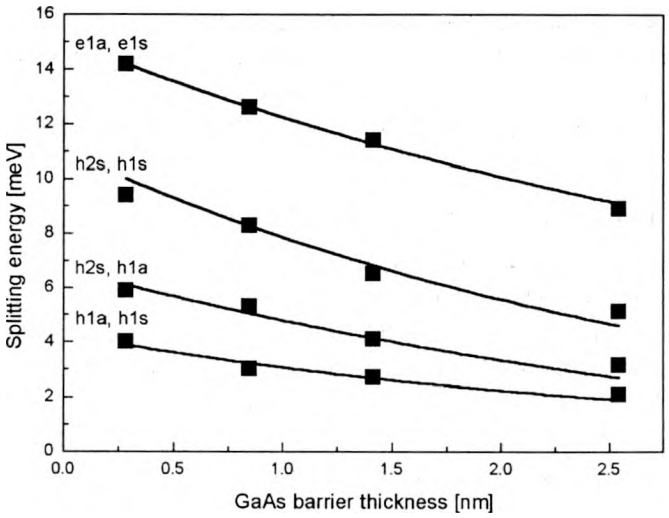

Fig. 6. Level splitting energy *vs.* GaAs barrier thickness for three coupled $In_{0.045}Ga_{0.955}As/GaAs$ quantum wells. Squares – experiment; solid lines – calculations.

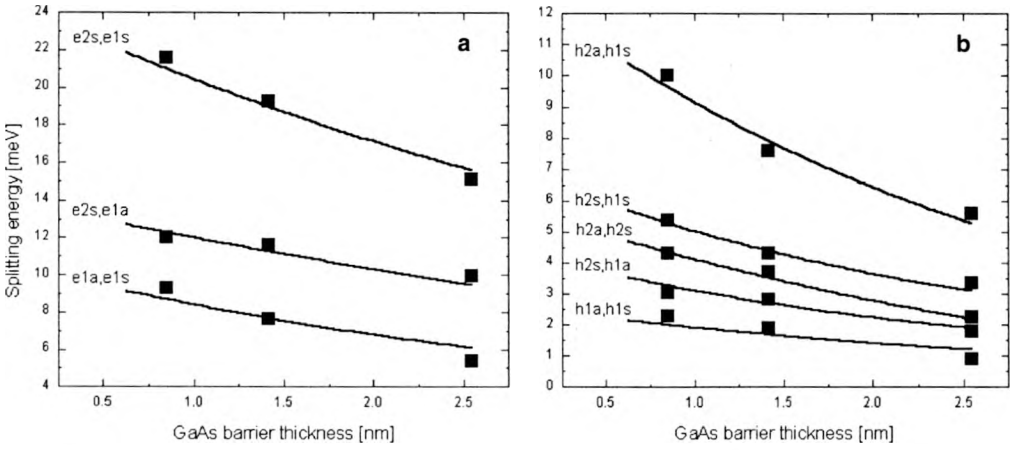

Fig. 7. Level splitting energy *vs.* GaAs barrier thickness for four coupled $In_{0.045}Ga_{0.955}As/GaAs$ quantum wells: for electrons (**a**) and for heavy holes (**b**). Squares – experiment; solid lines – calculations.

field significantly influences the intensity of the forbidden transitions in such double quantum well [7].

The observation of forbidden transitions allows determining the splitting energy of electron and heavy hole levels directly from the spectra, simply by taking the difference between the energy of forbidden transition and respective allowed transition, *e.g.*, e1–hh2 (hh1a) and e1–hh1 (hh1s) gives the splitting energy between two lowest heavy hole levels (symmetric and antisymmetric ones): hh2–hh1 (hh1a–hh1s). We have performed such an analysis for three and four coupled

$In_{0.045}Ga_{0.955}As/GaAs$ quantum well samples, for which PR spectra had been measured [8]. The results are shown in Figs. 6 and 7, as a function of splitting energy versus thickness of the GaAs separating barrier. The experimental values of the splitting have been compared with the calculated ones. In spite of the very small splitting energies a very good agreement has been obtained.

3.4. $In_{0.6}Ga_{0.4}As/GaAs$ quantum dot laser structure for 980 nm

Photoreflectance spectroscopy can be also successfully applied to the full device structure, like, *e.g.*, laser for 980 nm, where the active part consists of one layer of $In_{0.6}Ga_{0.4}As/GaAs$ quantum dots formed on a thin wetting layer. In Figure 8, a PR spectrum for such a sample measured at liquid nitrogen temperature is presented. There are a lot of transitions above the band gap energy of GaAs, occuring in complicated varying period superlattices system, cladding the waveguide and serving as Bragg reflectors for carriers to confine them in the active region. However, for us the most interesting is the quantum dot layer, for which the transitions are observed below

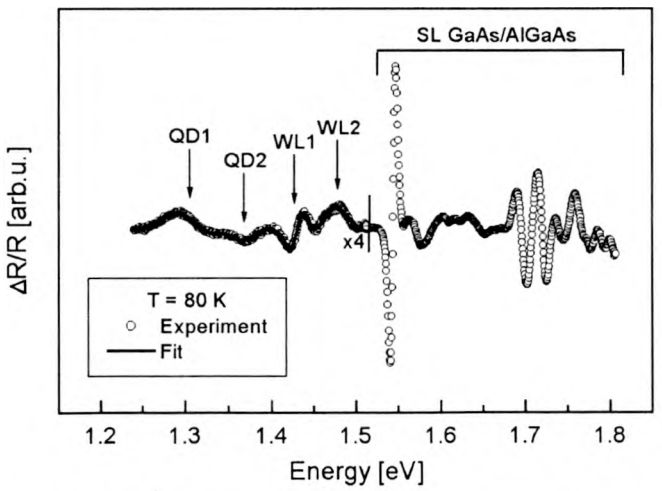

Fig. 8. PR spectrum of $In_{0.6}Ga_{0.4}As/GaAs$ quantum dot laser structure.

1.5 eV. We observe four transitions, two related to quantum dots (fundamental and excited one) and two related to wetting layer quantum well. Our calculations show that in this quantum well (4 ML thick) there is only one electron, one heavy hole and one light hole confined state. Hence, only two optical transitions are possible. The data from the literature show that for such quantum dots only two optical transitions are expected [9], [10].

3.5. Vertically coupled $In_{0.6}Ga_{0.4}As$ double quantum dots

In Figure 9, we show the PR spectra for coupled $In_{0.6}Ga_{0.4}As$ double quantum dot structures differing only in the thickness of GaAs separating layer [11]. The PR spectra

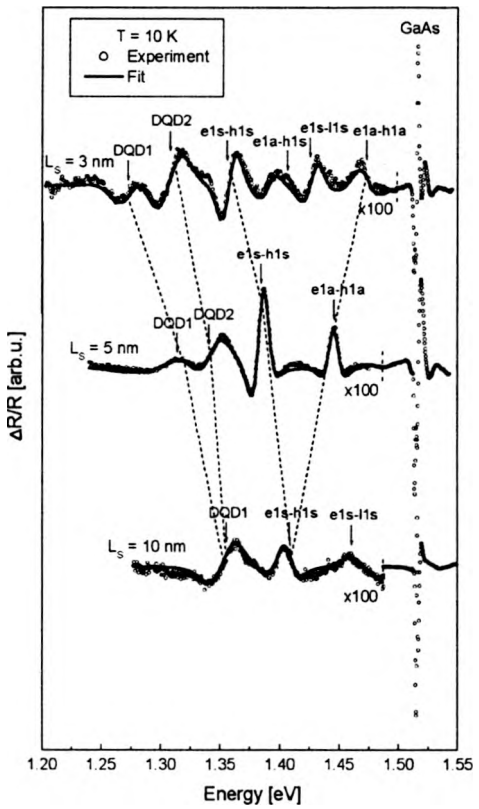

Fig. 9. Photoreflectance spectra of three DQD structures for various thicknesses of separating GaAs layer [11].

for all three samples can be divided into two parts. The strong feature at the 1.52 eV is related to the GaAs band gap transition. The low energy part of the spectra exhibits several transitions related to QDs and to the wetting layer. In order to identify the observed features we have calculated the energy levels in our double quantum dot and wetting layer double quantum well (WLDQW) (created from the two wetting layers) system. For DQWs we have used standard envelope function calculations including the strain effects. For the DQDs we have used the effective mass approximation for lens-shaped quantum dots that was previously developed by Wójs *et al.* [12]. Our dot is modelled by a part of a sphere formed on a thin wetting layer of thickness of 4 ML with a dot height and dot diameter of 2.5 and 18 nm, respectively, preserving the typical geometry of our indium content. The comparison of these theoretical results with the experimental ones obtained from the PR spectra is presented in Fig. 10 [11]. A good agreement has been obtained after including the exciton binding energy. For InGaAs quantum well transitions it should be about 8 meV. For excitons in InGaAs DQDs the binding energy is about 20 meV and depends on the indium concentration and dot shape.

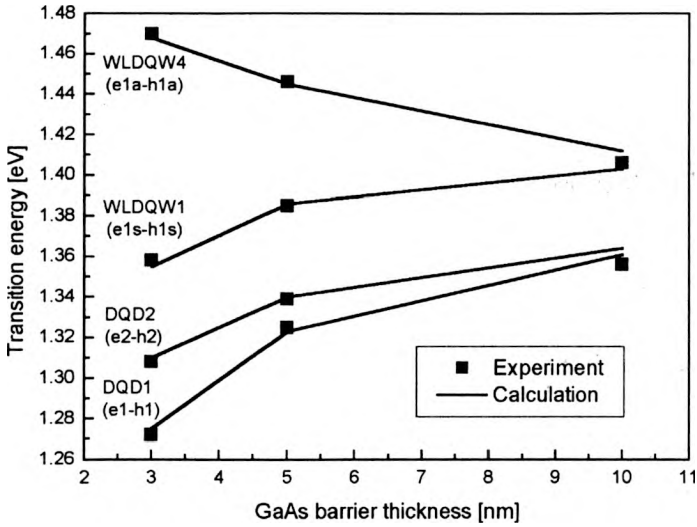

Fig. 10. Transition energy *vs.* GaAs separating layer thickness [11].

For the three various barrier thicknesses of GaAs layer, the strength of the electronic coupling between the dots and the wetting layer wells varies. For both, WLDQW and DQD systems the transitions splits, and the splitting decreases with increasing barrier width, demonstrating a strong dependence on the barrier width. In the case of 10 nm GaAs layer, no splitting has been observed showing that almost uncoupled case has been achieved.

4. Summary

Photoreflectance spectroscopy has been presented as a powerful technique for the non-destructive characterisation of low-dimensional semiconductor structures, especially those which can be applied as an active medium in novel semiconductor devices. This modulation technique usually allows to obtain more information about the sample at room temperature then common optical methods at low temperatures. The possibility of probing the excited state transitions, including those with little oscillator strength, belongs to the main advantages. It makes possible the detailed investigation of different kinds of structures like quantum wells, coupled quantum wells and quantum dots used in broad group of devices, from semiconductor lasers to quantum computer logical elements.

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