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Influence of Built-in Electric Field on Forbidden Transitions in $In_xGa_{1-x}As$/GaAs Double Quantum Well by Three-Beam Photoreflectance

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Photoreflectance spectroscopy has been used to study optical transitions in $In_{0.045}Ga_{0.955}As$/GaAs double quantum well at 80 K. The derivative nature of this contactless electromodulation technique allows for the observation of excited state transitions in the low-dimensional structure including the symmetry-forbidden ones. Excitonic symmetry-forbidden transitions can be observed due to the effect of mixing of heavy and light hole excitons and/or due to some asymmetry in the structure. We have shown that the built-in electric field in the region of double quantum well is weak enough (less than 0.5 kV/cm) not to cause any significant energetic shift of features due to quantum confined Stark effect, on one hand. On the other hand, it is sufficient to change strongly the oscillator strength of forbidden transitions. To change the internal electric field, we have used photoreflectance in the three-beam mode with a third beam continuously illuminating the sample and causing changes of the built-in electric fields due to the photovoltage effect. This method works as a contactless forward bias and allows for a change of the field down to the flat band conditions. We have shown that changes of built-in electric field by amount of a few tenths of kV/cm can modify the intensity of forbidden transitions significantly. We show that, although the mixing of excitons is still important, a very weak built-in electric field can be dominant in the observation of forbidden excitonic transitions in double quantum well.

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1. Introduction

Electronically symmetric coupled double quantum wells (CDQWs) are structures where two quantum wells are separated by a thin barrier layer. Both experiment and theory show that when the barrier is so narrow that there is a considerable overlap of wave functions in the two wells, the single quantum well electronic one-particle states split into symmetric and antisymmetric states with different energy levels [1, 2]. The splitting of energy levels is very sensitive to barrier widths and barrier heights and increases with decreasing barrier width and decreasing barrier height. Furthermore, the barrier controls also the importance of excitonic effects. In the case of narrow barriers, the single-particle splitting is large in comparison to the exciton binding energy. Then excitonic effects can be neglected to a good approximation. In other cases excitonic effects dominate the coupling and the Coulomb interaction mixes symmetric and antisymmetric single-particle states and generates intrawell and interwell exciton states.

Investigations on CDQWs have gained considerable interest during last decade. The interest in these structures 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 optoelectronic devices [3, 4]. Particular attention has been paid to the effect of a static electric field applied parallel to the structure growth axis [5–8]. There are also some papers studying the so-called parity-forbidden transitions in double quantum wells and showing that their appearance is mainly due to the valence band mixing effects causing the mixing of heavy and light hole excitons [9, 10].

In this work we show the results of photoreflectance (PR) spectroscopy applied to an $In_{0.045}Ga_{0.955}As/GaAs$ double quantum well structure. There are many papers demonstrating high sensitivity of photoreflectance, as a contactless form of electromodulation, to the investigation of coupled quantum wells, especially excited state transitions including symmetry-forbidden ones [11–13]. In the present work we focus our attention on a dependence of intensity of forbidden transitions on the built-in electric field via three-beam photoreflectance (TBPR) spectroscopy. In this mode of the PR experiment, the electric field in the region of CDQW is changed by varying the power density of additional (except probe and pump beams) laser illumination.

2. Experimental

Undoped $In_{0.045}Ga_{0.955}As/GaAs$ double quantum well structure was grown by solid-source molecular beam epitaxy on semi-insulating (001) GaAs substrate covered by 500 nm thick GaAs buffer. The width of wells was 7.5 nm and the separating GaAs barrier thickness was 9 monolayers (ML), where 1 ML corresponds to 0.28 nm. The whole structure was capped by 200 nm of GaAs.

Photoreflectance measurements were performed at 80 K where the sample was mounted on a cold finger of a continuous flow cryostat. Light from a halogen lamp dispersed through a double grating monochromator was used as a probe beam and 632.8 nm line of a He–Ne laser, chopped at frequency of 125 Hz, served as a pump beam. The power densities were 5 and 50 μW/cm^2, respectively. The reflected light was detected by a silicon photodiode. Third beam from a semiconductor laser (emitting at 645 nm) was used as a source of photoinduced changes of the built-in electric field, in the case of three-beam configuration of the experiment. Power density of the latter light was varied by neutral density filter. Further details of the experimental setup can be found in the literature [14–16].

3. Results and discussion

First, we calculated the energy levels of the single-particle states in the investigated double quantum well. We used standard envelope function approximation [17]. Due to the different lattice constants of In$_{0.045}$Ga$_{0.955}$As and GaAs the strain effects had to be included. Material parameters were taken from the literature [18, 19]. To obtain good agreement with the experiment, the energies of the optical transitions were corrected by binding energies of respective excitons [10]. It has been obtained that we have only two electron and two heavy hole confined states in the DQW: one symmetric and one antisymmetric for both types of carriers, which we denote as e 1 s,e 1 a and h 1 s,h 1 a for electrons and heavy holes, respectively. The light holes are unconfined in the GaAs barriers, so for light holes the system is of type II. Therefore, we can expect at most six optical transitions in the PR spectrum: four heavy hole-related (e 1 s–h 1 s, e 1 s–h 1 a, e 1 a–h 1 s, e 1 a –h 1 a), including the parity-forbidden ones, and perhaps two indirect in the real space light hole transitions (e 1 s–l, e 1 a–l). The calculated splitting between states is large enough to neglect the mixing of symmetric and antisymmetric states, in a first approximation.

Figure 1 shows PR spectrum of investigated sample at 80 K. Indeed, six features are observed below the GaAs bulk-like related signal. The experimental transition energies have been obtained from the fitting procedure according to first derivative Gaussian lineshape (FDGL), the most appropriate form of PR signal in the case of excitonic transitions [14–16]. The transitions have been identified basing on the results of the calculations. For us, the most interesting is the region of the forbidden transitions, i.e. transitions between the states of different symmetry (e 1 s–h 1 a and e 1 a–h 1 s). It was shown earlier that strong mixing of heavy and light hole excitons is responsible for the observation of these transitions in such a structure. However, we suppose that, although the DQW is about 200 nm from the surface, it can be still in a weak electric field due to the band bending at the surface. The field can be assumed to be homogeneous in the narrow region of quantum well. Therefore, we performed the TBPR experiment for this sample

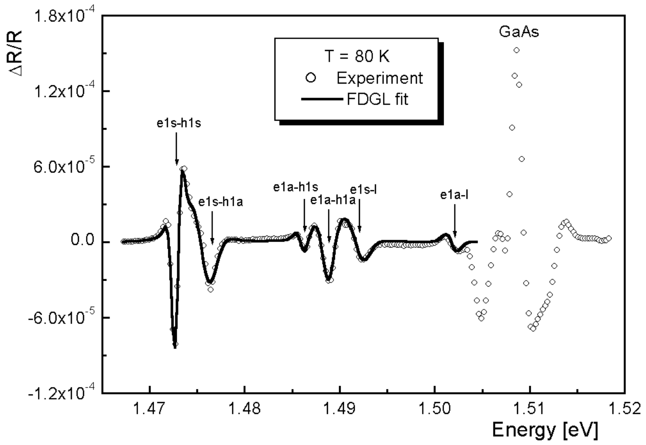

Fig. 1. Photoreflectance spectrum of $In_{0.045}Ga_{0.955}As/GaAs$ double quantum well structure.

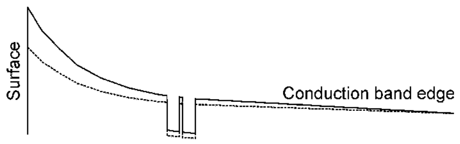

Fig. 2. Sketch of the conduction band edge at the surface of structure with double quantum well. The influence of the above band gap illumination is shown by dashed line.

using an additional light from a laser with energy higher than the GaAs band gap, to change the built-in electric field due to the photovoltage effect. A sketch of such a light-induced changes in the band bending of a structure with double quantum well is shown in Fig. 2. The existence of the built-in electric field in the region of DQW destroys the symmetry of the system and therefore can give its own contribution to the forbidden transition oscillator strength. If we change the field, the overlap integrals and hence the intensity of these transitions are also changed.

Figure 3 shows a comparison of the PR spectrum from Fig. 1, in the region of fundamental allowed and those two discussed forbidden transitions, with TBPR spectrum measured for above 100 $\mu W/cm^2$ power density of the third beam. The decomposition of this part of the spectra into single PR lines is shown in Fig. 4, for various power densities. It is clearly seen that the intensity of the allowed transition (e 1 s–h 1 s) is almost unchanged, whereas the intensity of the forbidden transition decreases significantly under influence of additional illumination. It is also worth noticing that no energetic shifts are observed. The changes of the built-in electric field are weak enough not to cause any shifts of the levels due to quantum confined Stark effect, but they are sufficient to influence the intensity

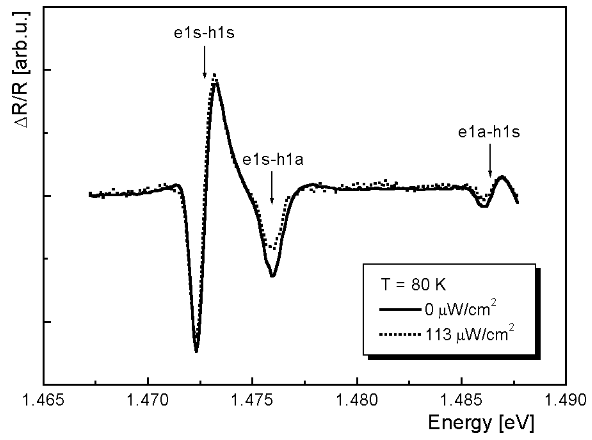

Fig. 3. Comparison of PR and TBPR spectra for $In_{0.045}Ga_{0.955}As$/GaAs double quantum well in the region of forbidden transitions.

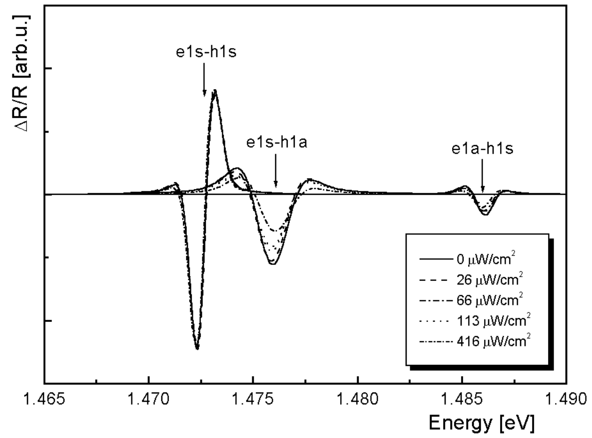

Fig. 4. Decomposition of TBPR spectra measured at various third beam power densities into single photoreflectance features.

of the forbidden transitions. The lack of the energy shift of PR features under additional illuminations allow us to neglect many-body effects and effects related to the filling of the lowest subband, suggesting that we are still in the regime of low concentrations of carriers in the well.

The dependence of the ratio of the intensity of forbidden transitions to the intensity of the fundamental allowed one on the third beam power density is shown in Fig. 5. This ratio decreases with the increase in the power density, which is equivalent to the decrease in the built-in electric field. This dependence seems to saturate for higher power densities suggesting that we are close to the flat band conditions in the region of DQW. Even then no Stark shifts of transitions are observed, therefore we can say that the built-in electric field in the unilluminated sample has to be very weak. Our calculations show that the levels start to shift significantly (more than 0.1 meV) for fields higher than 0.5 kV/cm. Additionally, the electric field higher than 0.5 kV/cm cause strong leakage of the wave function of the antisymmetric state of electron, because the energy of this state is close to the energy of the conduction band edge of the GaAs barrier. The electric field parallel to the growth direction tilts the bands (see Fig. 2), so that the electron starts to escape from the well through the triangle barrier due to the tunnelling. It should significantly decrease the intensity of transitions (also allowed ones) involving this e 1 a state. Hence, we can conclude that the built-in electric field in the region of double quantum well is at most of an order of 0.5 kV/cm. In the TBPR experiment we can only decrease this field by amount of several tenths of kV/cm. Therefore, we can not produce any Stark shifts of the PR features, but we can cause only the changes of the intensity, especially for the forbidden transitions.

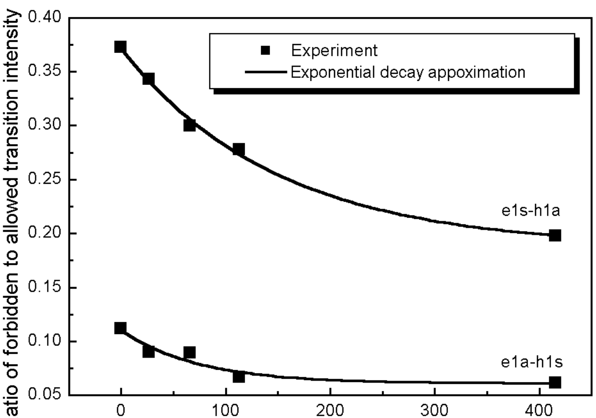

Fig. 5. Third beam power density dependence of the ratio of forbidden to fundamental allowed transition intensities obtained from TBPR spectra.

Usually, the built-in electric field depends approximately linearly on the logarithm of the power density of the sample illumination [20], especially in a relatively narrow range of power density changes. Therefore, the dependence shown in Fig. 5, plotted as a function of the third beam power density in the logarithmic scale reflects the dependence on the built-in electric field. Such a function is presented in Fig. 6. For both forbidden transitions this dependence can be approximated by a straight line. To confirm this tendency, at least qualitatively, we show also (in the inset of Fig. 6) the dependence of the calculated overlap integrals ratio of forbidden e 1 s–h 1 a and allowed e 1 s–h 1 s transitions, as a function of the electric fields of about 0.5 kV/cm. This is really an almost linear function in the range of a few tenths of kV/cm. It has to be mentioned that the absolute value of this ratio

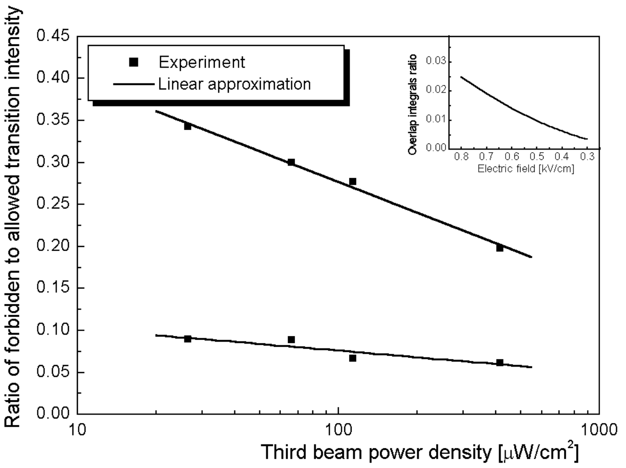

Fig. 6. Dependence of the ratio of forbidden to fundamental allowed transition intensities on the third beam power density in the logarithmic scale. The inset shows the electric field dependence of the calculated ratio of overlap integrals of forbidden (e 1 s–h 1 a) and allowed (e 1 s–h 1 s) transitions.

is about an order of magnitude lower than the intensity ratio from TBPR. This is probably due to the simplicity of calculations, which do not include excitonic effects explicitly. Especially, the influence of mixing effects of heavy and light hole excitons as well as the possible mixing of symmetric and antisymmetric states can be important in the correct determination of the oscillator strength of forbidden transitions. Therefore, our calculations are not able to explain such a high experimental value of the intensity ratio shown in Figs. 5 and 6, neither the difference in the power density dependence for both observed forbidden transitions.

4. Conclusions

In summary, we have measured photoreflectance spectroscopy on the $\mathrm{In}_{0.045}\mathrm{Ga}_{0.955}\mathrm{As/GaAs}$ double quantum well structure. We have observed all the possible optical confined state-related transitions in this structure including indirect in the real space light hole transitions and parity-forbidden heavy hole transitions. We have investigated the dependence of the intensity of the forbidden transitions on the built-in electric field by using a third light beam in PR experiment. The ratio of the intensity of forbidden transitions to the intensity of fundamental allowed transition depends linearly on the third beam power density and hence approximately linearly on the internal electric field in the region of DQW. This result has been qualitatively confirmed by simple calculations of the dependence of the overlap integrals of those transitions as a function of weak, parallel to the growth axis, electric field. The conclusion is that even very weak built-in electric field in the double quantum well is also important, except exciton mixing effects, in the observation of symmetry forbidden transitions.

Acknowledgments

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References

[1] T. Kamizato, M. Matsuura, *Phys. Rev. B* **40,** 8378 (1989).

[2] M. Bayer, V.B. Timofeev, F. Faller, T. Gutbrod, A. Forchel, *Phys. Rev. B* **54,** 8799 (1996).

[3] M.N. Islam, R.L. Hillman, D.A.B. Miller, D.S. Chemla, A.C. Gossard, J.H. English, *Appl. Phys. Lett.* **50,** 1098 (1987).

[4] Y. Tokuda, K. Kanamoto, N. Tsukada, *Appl. Phys. Lett.* **56,** 166 (1990).

[5] Y.J. Chen, E.S. Koteles, B.S. Elman, C.A. Armiento, *Phys. Rev. B* **36,** 4562 (1987).

[6] S.R. Andrews, C.M. Murray, A.A. Davies, T.M. Kerr, *Phys. Rev. B* **37,** 8198 (1988).

[7] I. Galbraith, G. Duggan, *Phys. Rev. B* **40,** 5515 (1989).

[8] M.M. Dignam, J.E. Sipe, *Phys. Rev. B* **43,** 4084 (1991).

[9] T. Wang, M. Bayer, A. Forchel, *Phys. Rev. B* **58,** R10183 (1998).

[10] T. Wang, M. Bayer, A. Forchel, N.A. Gippius, V. Kulakovskii, *Phys. Rev. B* **62,** 7433 (2000).

[11] G. Sęk, K. Ryczko, M. Kubisa, J. Misiewicz, J. Koeth, A. Forchel, *Opto-electronics Rev.* **7,** 117 (1999).

[12] G. Sęk, K. Ryczko, M. Ciorga, L. Bryja, M. Kubisa, J. Misiewicz, M. Bayer, J. Koeth, A. Forchel, in: *Proc. of Advanced Research NATO Workshop on Optical Properties of Semiconductor Nanostructures*, Eds. M.L. Sadowski, M. Potemski, M. Grynberg, Kluwer Academic Publishers, Dordrecht 2000, p. 91.

[13] G. Sęk, K. Ryczko, M. Kubisa, J. Misiewicz, M. Bayer, T. Wang, J. Koeth, A. Forchel, *Thin Solid Films* **364,** 220 (2000).

[14] J. Misiewicz, P. Sitarek, G. Sęk, *Introduction to the Photoreflectance Spectroscopy of Semiconductor Structures*, Oficyna Wydawnicza Politechniki Wrocławskiej, Wrocław 1999, p. 19.

[15] J. Misiewicz, G. Sęk, P. Sitarek, *Optica Applicata* **39,** 327 (1999).

[16] J. Misiewicz, P. Sitarek, G. Sęk, *Opto-electronics Rev.* **8,** 1 (2000).

[17] G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures*, Les Editions de Physique, Les Ulis 1990.

[18] G. Arnaud, J. Allegre, P. Lefebvre, H. Mathieu, L.K. Howard, D.J. Dunstan, *Phys. Rev. B* **46,** 15290 (1992).

[19] L.C. Andreani, D. De Nova, S. Di Lernia, M. Geddo, G. Guizzetti, M. Patrini, C. Bocchi, A. Bosacchi, C. Ferrari, S. Franchi, *J. Appl. Phys.* **78,** 6745 (1995).

[20] H. Shen, M. Dutta, *J. Appl. Phys.* **78,** 2151 (1995).