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Optical absorption in GaAs quantum wells caused by donor–acceptor pair transitions

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Abstract

Optical absorption related to donor–acceptor pair transitions in GaAs quantum wells was theoretically investigated. The donor and acceptor ground state wavefunctions and energies were obtained using variational method in the frames of the effective mass approximation. The absorption coefficient related to acceptor-to-donor transition was calculated. The spreading of the absorption curve as a result of averaging over impurity distances was taken into account. The character of the dependence of the absorption coefficient on both quantum well thickness and impurity concentration was determined. The blueshift in the absorption spectra was theoretically investigated. Comparison with available experimental data was made.

1. Introduction

The presence of doping impurity centres in semiconductor quantum well (QW) structures has a significant influence on their properties. The investigation of properties related to these impurities is not only of fundamental interest, but is also of major importance in optoelectronic device applications (high electron mobility transistors, QW infrared photodetectors or emitters, etc) [1, 2].

The optical properties of doped bulk semiconductors have been intensively studied. In semiconductors of the A^3B^5 group, the electrons that bind with doped impurities form shallow energy states close to band gap edges [3]. In particular, doping the bulk semiconductor simultaneously with both donor and acceptor impurities forms shallow states near the band gap edges, making possible optical transitions between two clearly isolated energy levels [4–11].

One of the first works related to investigations of donor–acceptor pair (DAP) transitions in the above-mentioned semiconductors was that of Hopfield [4]. Later, detailed investigations of DAP transitions were made by Stoneham and Harker [5, 6], where central-cell corrections

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were taken into account. In particular, in [6] the dependence of the transition energy on average distance R between DAP in GaP:(S, C) for different levels of calculations was examined in detail.

Significant study of impurity optical properties in QW structures was performed in [12–23]. A detailed investigation of hydrogenic impurities in GaAs QW was performed by Bastard [12]. Later, investigations of impurity properties in QW were followed by several other more detailed investigations [13–17]. The influence of mismatches of the dielectric constant at well interfaces was also considered [14]. The effects of spatially dependent screening, electron–phonon interactions, nonparabolicity of the conduction and valence band, etc were considered by other authors [15–17]. The energy spectrum of the ground state and the low-lying excited states for shallow impurities in QW structures was studied [18, 19]. Observations of impurity related photoluminescence (PL) features were reported in [21, 22].

Developments in experimental techniques led to increased interest in the possibilities of the regulation of the optical features of QWs by means of external magnetic fields [18, 24–26] and also with consideration of polaron effects [26].

Variations in the properties can be caused by changing the concentration of the dopant from a uniform distribution within the QW to concentrated sheet layers, resulting in a so-called δ-doped profile. It is possible to tune the energetic levels of an impurity in a controlled way by changing the doping profile. Understanding the influence of impurities on the optical properties near the QW intrinsic transitions is of particular concern in order to optimize the design of optoelectronic devices.

The PL spectrum is an effective technique for characterizing doped QWs. In addition to free-excitonic transitions, donor-bound exciton [27, 28], acceptor-bound exciton [29], free electron to acceptor [20, 30, 31] and heavy hole to donor [20, 30, 31] transitions were also observed in p-type and n-type doped QWs.

Besides the above-mentioned ones, acceptor-to-donor pair transitions in QWs are also possible. After a careful literature search we found several experimental reports concerning the observation of a donor–acceptor pair transition peak in the PL spectra. Uchida *et al* [32] have investigated the so-called '1.46 eV deep emission band' observed in the PL spectra of 100 Å thick $Ga_{0.52}In_{0.48}P/GaAs$ single QWs, grown by metal organic vapour phase epitaxy (MOVPE). Based on the pressure dependence results, they suggested that the 'deep emission' peak is related to DAP transitions. However, later in [33, 34] using a low temperature, they suggested that the 1.46 eV emission is a spatially indirect transition of the electrons and holes, separated at the interface in a type-II band alignment. Nevertheless, at the same time Ding *et al* [35] reported the observation of an anomalously large blueshift of an apparent DAP transition peak in compensation-doped coupled QWs. The blueshift was observed in PL spectra while the excitation intensity increases from 0.54 to 423 W cm^{-2}. The authors proposed that the blueshift is due to the change of the Coulomb interaction energy between recombined donors and acceptors as their separation decreases. Later Guzman *et al* [36] performed an optical characterization of GaAs/GaAlAs single QW structures by interband PL spectroscopy. The peak in the PL spectra at lower energy was observed and attributed to a DAP transition. Samples were grown by molecular beam epitaxy (MBE) with a two-dimensional doping concentration (Si) in the wells in the range of $0–10^{12}$ cm^{-2}. Si is related to the acceptor, while C is related to the residual donor, which is always present in samples grown by MBE. In this connection, one can assume that non-compensated QWs were considered. The dependence of the DAP transition peak on different doping concentrations was examined, and the blueshift was observed.

However, in the above-mentioned experimental works, discussions about DAP transitions concerned only qualitative aspects of the subject. In this connection, it is important to have

a quantitative model, which will describe the aforementioned transitions, that can give the opportunity to perform an essential comparison between theory and the experiment.

In this paper we present a theoretical investigation of DAP transitions in the framework of non-compensated lightly doped GaAs infinite-barrier QWs.

2. Theory

The impurity envelope functions are the solutions of the Schrödinger equation with the effective Hamiltonian

$$\hat{H} = \frac{\hat{p}^2}{2m^*} + V(z) - \frac{e^2}{\kappa\sqrt{\rho^2 + (z - z_i)^2}}, \tag{1}$$

where m^* is the electron effective mass, κ is the dielectric constant (for GaAs $\kappa = 13.18$), z_i ($i = \mathrm{D}, \mathrm{A}$) is the impurity position along OZ axes and $V(z)$ is the confinement potential. We consider a rectangular infinite-barrier QW

$$V(z) = \begin{cases} 0, & |z| < \frac{L}{2} \\ \infty, & |z| \geqslant \frac{L}{2}. \end{cases}$$

For definiteness we will only refer to the donor state, because it is clear that (1) also applies to the acceptor state, where m^* is understood as the hole effective mass.

We present the envelope function of the ground state as

$$\psi_0(\rho, z) = \varphi_0(\rho)\chi_0(z), \tag{2}$$

where $\varphi_0(\rho)$ is the function in the QW plane and $\chi_0(z)$ is along the quantization axis OZ. Thus to define the ground state of the considered system we have the Schrödinger equation in the form

$$\left[\frac{\hat{p}_x^2 + \hat{p}_y^2}{2m^*} + \frac{\hat{p}_z^2}{2m^*}\right]\varphi_0(\rho)\chi_0(z) + V(z)\varphi_0(\rho)\chi_0(z) - \frac{e^2}{\kappa}\frac{\varphi_0(\rho)\chi_0(z)}{\sqrt{\rho^2 + (z - z_i)^2}} = \varepsilon\varphi_0(\rho)\chi_0(z). \tag{3}$$

Taking into consideration the following:

$$\frac{\hat{p}_z^2}{2m^*}\chi_0(z) + V(z)\chi_0(z) = E_0\chi_0(z), \tag{4}$$

where $E_0 = \frac{\pi^2\hbar^2}{2m^*L^2}$, then we multiply both sides of (3) by $\chi_0^*(z)$ and integrate over z between $-L/2$ and $L/2$. Taking into account the normalization condition for $\chi_0(z)$ we get a two-dimensional Schrödinger equation for the function $\varphi_0(\rho)$

$$\left[\frac{\hat{p}_x^2 + \hat{p}_y^2}{2m^*} + V_{\mathrm{eff}}(\rho)\right]\varphi_0(\rho) = (\varepsilon - E_0)\varphi_0(\rho), \tag{5}$$

where $V_{\mathrm{eff}}(\rho)$ is the effective Coulomb potential in the XOY plane:

$$V_{\mathrm{eff}}(\rho) = -\frac{e^2}{\kappa}\int_{-\frac{L}{2}}^{\frac{L}{2}} \frac{|\chi_0|^2}{\sqrt{\rho^2 + (z - z_i)^2}}\,\mathrm{d}z. \tag{6}$$

The solution of equation (5) is found using the variational method, with the trial function of the ground state in the form [38]

$$\varphi_0(\rho) = \frac{1}{\lambda}\sqrt{\frac{2}{\pi}}\mathrm{e}^{-\rho/\lambda}, \tag{7}$$

where λ is the variational parameter.

The ground state energy is obtained after minimization of the function

$$\varepsilon(\lambda, z_i) = E_0 + \frac{\hbar^2}{2m^*\lambda^2} - \frac{2e^2}{\kappa\lambda}\int_0^\infty x\,\mathrm{e}^{-x}\,\mathrm{d}x\int_{-\frac{L}{2}}^{\frac{L}{2}}\frac{|\chi_0|^2\,\mathrm{d}z}{\sqrt{x^2+\frac{4}{\lambda^2}(z-z_i)^2}}. \tag{8}$$

The binding energy is equal to

$$E_{\mathrm{bind}} = E_0 - \min_{\lambda}\varepsilon(\lambda, z_i). \tag{9}$$

Now we turn to the calculation of the absorption coefficient in the considered structure, conditioned by transitions between the ground states of the DAP.

Let us consider a lightly doped QW with a concentration of acceptors n_{A}, so that $\bar{R} \gg a_{\mathrm{D}}, a_{\mathrm{A}}$ conditions are satisfied ($\bar{R}$ is the average distance between acceptors and donors in the QW plane). In this case the main contribution to the transitions within the donor–acceptor system make pairs satisfying the conditions $R \geqslant a_{\mathrm{D}}, a_{\mathrm{A}}$, because the number of pairs with $R < a_{\mathrm{D}}$ is not significant. For this case the coupling energy of the DAP can be taken equal to $e^2/\kappa R$ and considered as an acceptor energy level shift. This case corresponds to the Coulombic approximation mentioned in [6], which means that in this work we do not refer to more general central-cell correction approach [6]. The location of the donor is $(\vec{\rho}, z)$, and that of the acceptor is $(\vec{\rho}-\vec{R}, z)$, where $\vec{\rho}$ is the radius vector and $\vec{R}$ is the distance between the donor and acceptor in the QW plane. Later we will assume that the donor and acceptor are located in the centre of a QW.

The electron and hole ground eigenstates and eigenvalues are (measured from the maximum of valence band)

$$\psi_{\mathrm{A}} = \frac{2}{\lambda_{\mathrm{A}}}\sqrt{\frac{1}{\pi L}}\cos\left(\frac{\pi z}{L}\right)\mathrm{e}^{-\frac{|\vec{\rho}-\vec{R}|}{\lambda_{\mathrm{A}}}}u_{v,0}(\vec{\rho}-\vec{R}), \tag{10}$$

$$\psi_{\mathrm{D}} = \frac{2}{\lambda_{\mathrm{D}}}\sqrt{\frac{1}{\pi L}}\cos\left(\frac{\pi z}{L}\right)\mathrm{e}^{-\frac{\rho}{\lambda_{\mathrm{D}}}}u_{c,0}(\vec{\rho}), \tag{11}$$

$$E_{\mathrm{A}} = -\min\varepsilon_{\mathrm{A}}(\lambda_{\mathrm{A}}) - \frac{e^2}{\kappa R}, \tag{12}$$

$$E_{\mathrm{D}} = \min\varepsilon_{\mathrm{D}}(\lambda_{\mathrm{D}}) + \varepsilon_{\mathrm{gap}} \tag{13}$$

where λ_{A}, λ_{D} are variational parameters and $u_{v,0}$, $u_{c,0}$ are Bloch amplitudes in the centre of the Brillouin zone (in the discussed structure zone extrema are on the centre of Brillouin zone).

The light absorption coefficient is determined by the formula [37]

$$\alpha_R(\omega) = \frac{4\pi^2 c}{N\omega V}\frac{|M_{\mathrm{AD}}|^2}{|A_0|^2}\delta\left(E_f - E_i - \hbar\omega\right), \tag{14}$$

where V is the sample volume, M_{AD} is the matrix element of the 'acceptor → donor' transition, N is the refractive index and A_0 is the vector potential amplitude of the incident electromagnetic wave.

In the case of normally incident light the matrix element can be written as

$$\begin{aligned} M_{\mathrm{AD}} &= \frac{2ec}{\pi m_0 c}(\vec{e}\vec{p}_{cv})\frac{1}{\lambda_{\mathrm{A}}\lambda_{\mathrm{D}}}\frac{2}{L}\int_{-\frac{L}{2}}^{\frac{L}{2}}\mathrm{e}^{\mathrm{i}q_z z}\cos\left(\frac{\pi z}{L}\right)\cos\left(\frac{\pi z}{L}\right)\mathrm{d}z \\ &\quad\times\int_0^{2\pi}\int_0^\infty \mathrm{e}^{-\left(\frac{1}{\lambda_{\mathrm{A}}}\sqrt{\rho^2+R^2-2\rho R\cos\varphi}+\frac{1}{\lambda_{\mathrm{D}}}\rho\right)}\rho\,\mathrm{d}\rho\,\mathrm{d}\varphi \\ &= \frac{2ec}{\pi m_0 c}(\vec{e}\vec{p}_{cv})\frac{1}{\lambda_{\mathrm{A}}\lambda_{\mathrm{D}}}F(R)\xi(q_z L) \end{aligned} \tag{15}$$

where $\vec{p}_{cv}$ is the matrix element conditioned by Bloch amplitudes, $\vec{e}$ is the incident light polarization and q_z is the photon wavevector in the z direction.

By $F(R)$ and $\xi(q_z L)$ we denote the following integrals

$$F(R) = \int_0^{2\pi} \int_0^{\infty} \mathrm{e}^{-\left(\frac{1}{\lambda_\mathrm{A}}\sqrt{\rho^2+R^2-2\rho R\cos\varphi}+\frac{1}{\lambda_\mathrm{D}}\rho\right)} \rho \,\mathrm{d}\rho \,\mathrm{d}\varphi, \tag{16}$$

$$\xi(q_z L) = \frac{2}{L}\int_{-\frac{L}{2}}^{\frac{L}{2}} \mathrm{e}^{\mathrm{i}q_z z} \cos\left(\frac{\pi z}{L}\right) \cos\left(\frac{\pi z}{L}\right) \mathrm{d}z. \tag{17}$$

In discussed case of shallow impurities equation (15) can be simplified if we take into consideration the fact that for GaAs $\tilde{\varepsilon}_g \lesssim \varepsilon_g \sim 1$ eV (the expression for $\tilde{\varepsilon}_g$ see below) and for the QW width we have $L \sim 10^{-6}$ cm (∼100 Å), which makes the parameter $q_z L \ll 1$. This allows us to take $\xi(q_z L)$ approximately equal to 1.

As follows from equation (14), such transitions are possible only between those DAPs the distances between which can be determined from the energy conservation law

$$R_1 \equiv R = \frac{e^2}{\kappa(\hbar\omega - \tilde{\varepsilon}_g)} \tag{18}$$

where

$$\tilde{\varepsilon}_g = \varepsilon_g + E_\mathrm{D}^0 + E_\mathrm{A}^0, \qquad E_\mathrm{A}^0 = \min_{\lambda_\mathrm{A}} \varepsilon_\mathrm{A}(\lambda_\mathrm{A}), \qquad E_\mathrm{D}^0 = \min_{\lambda_\mathrm{D}} \varepsilon_\mathrm{D}(\lambda_\mathrm{D}). \tag{19}$$

Considering R as changing continuously when $n_\mathrm{A} \gg n_\mathrm{D}$, let us write the expression for the absorption coefficient [8]

$$\alpha(\omega) = N_\mathrm{D} \int_0^{\infty} \alpha_R(\omega) W(R) \,\mathrm{d}R \tag{20}$$

where N_D is the number of donors and $W(R)$ the distribution function by R values.

As the DAP distribution function we take the adjacent-neighbour distribution [39–41], and taking $W(R)$ not dependent on z:

$$W(R) = 2\pi R n_\mathrm{A} \exp\{-\pi R^2 L n_\mathrm{A}\} \tag{21}$$

where n_A is the bulk concentration of acceptors.

After averaging over the distribution (21) for the absorption coefficient of the DAP transition we obtain the following expression:

$$\alpha_\mathrm{DAP}(\omega) = \frac{2^5 \pi N n_\mathrm{A}^- n_\mathrm{D}^+}{L\omega\lambda_\mathrm{A}^2\lambda_\mathrm{D}^2 c m_0^2} |\vec{e}\vec{p}_{cv}|^2 |\xi\,(q_z L)|^2 |F(R_1)|^2 R_1^3 \exp\{-\pi R_1^2 n_\mathrm{A}^-\} \tag{22}$$

where n_A^- and n_D^+ are two-dimensional surface concentrations of acceptors and donors, respectively.

3. Discussion

Figure 1 shows the dependences of the absorption coefficient on the energy of incident light at different values of QW width ($L = 50$, 65, 100 Å). As follows from the figure, with the increase in L the effective width of the forbidden band decreases and therefore the absorption threshold shifts to the smaller energies (smaller frequencies) as a result of weakening of the size quantization. Also, a small reduction is observed in the value of the absorption coefficient. Calculations are made of the value of dominant impurity (acceptors) concentration, $n_\mathrm{A}^- = 10^{11}$ cm^{-2}, and at the $k = 0.05$ compensation value ($k = n_\mathrm{D}^+/n_\mathrm{A}^-$). The numerical values

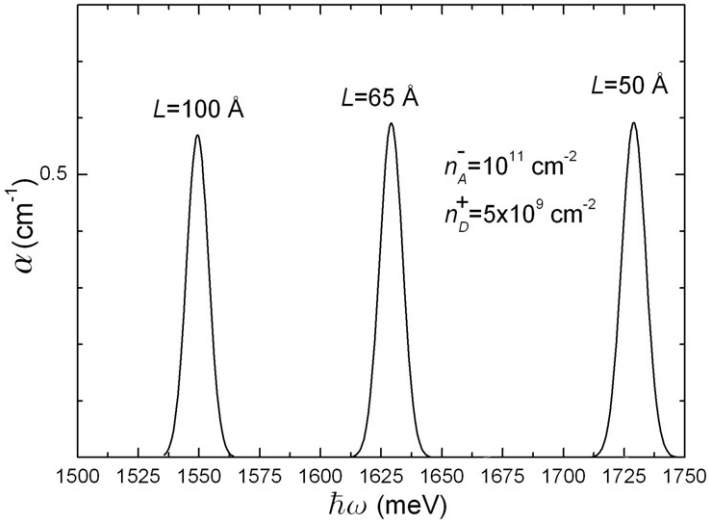

Figure 1. Dependence of the absorption coefficient on incident light frequency at different QW widths.

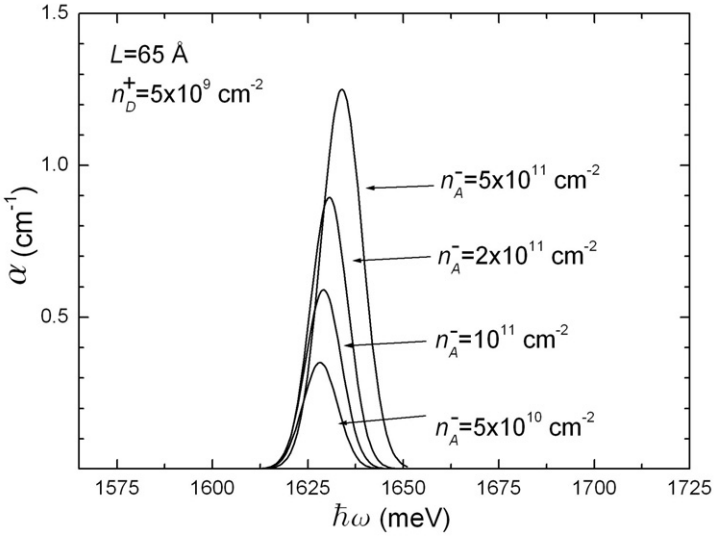

Figure 2. Dependence of the absorption coefficient on incident light frequency at different concentrations of dominant impurity.

of parameters in the absorption coefficient (22) in the GaAs structure are: $\varepsilon_{\text{gap}} = 1.519$ eV, $m_v = 0.34m_e$, $m_c = 0.067m_e$, $N = 3.6$.

Figure 2 presents the dependence of the absorption coefficient on the incident light frequency at different values of dominant impurity concentration ($n_A^- = 5 \times 10^{10}$ cm^{-2}, $n_A^- = 10^{11}$ cm^{-2}, $n_A^- = 2 \times 10^{11}$ cm^{-2}, $n_A^- = 5 \times 10^{11}$ cm^{-2}). With increase in the impurity concentration the absorption coefficient increases, and the threshold frequency slightly increases also (i.e. the blueshift is observable).

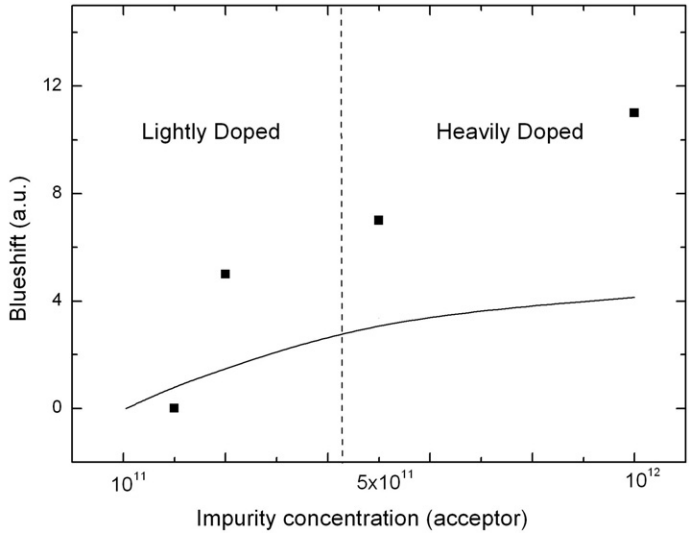

Figure 3. The dependence of the blueshift on dominant impurity (acceptor) concentration: squares, experimental results [36]; solid line, result of calculations; left, region of low concentration; right, region of high concentration.

Figure 3 shows the dependence of the blueshift on different values of two-dimensional concentrations of dominant impurity (acceptor). The difference between acceptor and donor energy levels can be presented as

$$E_{\mathrm{DAP}} = \varepsilon_g - E_{\mathrm{bD}} - E_{\mathrm{bA}} + \frac{e^2}{R}, \tag{23}$$

where E_{bD} and E_{bA} are donor and acceptor binding energies, respectively; the fourth item in equation (23) is the Coulombic term.

In the frames of our calculations the blueshift $\Delta E_{\mathrm{blueshift}}$ is proportional to the Coulombic term, i.e. inversely proportional to the distance between donor and acceptor

$$\Delta E_{\mathrm{blueshift}} \sim \frac{e^2}{R}. \tag{24}$$

When the acceptor concentration is increasing (e.g. the concentration of Si atoms [36]), donors (e.g. residual C atoms [36]) and acceptors become spatially closer, the blueshift in the acceptor–donor transition peak should take place as a result of the increase in the Coulombic term [35, 36]. So the growth of doping level should be the reason for the increase in the blueshift. Such a result is obtained in our theoretical model. On the other hand there is a significant difference between the experimental and theoretical data for the lightly doped samples, related to the following reasons:

(1) It is reasonable to point out that the impurities in the GaAs studied in the frames of our model are assumed to be located in the centre of a QW. However, in experiment [36] samples have the same two-dimensional doping concentration as in the presented model, although the extension of the doping is 25–45 Å. As one can see from the experimental data [36] with the increase of dominant impurity concentration the doping extension increases. Thus, the relative number of acceptors near to the GaAs QW edge increases with the increase in doping concentration. The impurity binding energy decreases when the impurity location moves from the centre to the edge of the QW [12, 38]. The estimated

difference in acceptor binding energy between the theoretical and experimentally measured one, where acceptors are assumed to be randomly distributed in the GaAs wells, is about ∼4 meV for a 65 Å QW [42]. So it is reasonable to expect the divergence of the presented theoretical model from available experimental data.

(2) In the frames of our model we consider isolated impurity centres. Therefore, as was mentioned above, the Coulombic approximation of the more general case of central-cell correction was considered [6], and inter-impurity interactions are not discussed.

(3) In [36] the blueshift of the DAP transition peak was observed with respect to the e1–hh1 peak (transition between the first conduction subband and the first heavy-hole subband) in GaAs/AlGaAs QW infrared detector structures. The considered samples with different concentrations of impurities were grown during different runs via MBE. In the reported results fluctuations in QW thickness (appearing due to different runs) introduce a certain variation into the growth tendency of the blueshift along with increase in the impurity concentration in the lightly doped samples. The comparison of our theoretical results with the experimental data shows that in the lightly doped samples the mentioned technological fluctuations significantly affect the growth tendency of the blueshift along with the increase in impurity concentration from sample to sample. As it is seen from figure 1, even small differences in well thickness can result in significant shift of the absorption threshold (peak position).

4. Conclusion

We have presented a theoretical model for donor–acceptor pair transitions in non-compensation doped GaAs QWs. These transitions are taking place between the levels of different impurity atoms. The developed model enables us, in the frame of a simple theoretical model, to simulate the blueshift behaviour in doped QW structures.

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