OPTICAL SPECTRA IN THE REGION OF EXCITONIC RESONANCES IN QUANTUM WELLS AND QUANTUM DOTS OF In_{0.3}Ga_{0.7}As/GaAs HETEROSTRUCTURES

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Abstract. Lines conditioned by the transitions hh, lh1-e1(1s, 2s, 3s), hh2, lh2-e2(1s, 2s, 3s), hh1, lh1-e2(1s) and hh3, lh3-e3(1s) had been revealed in reflection spectra of $In_{0.3}$ $Ga_{0.7}As / GaAs$ nanostructures. The shapes of the reflection and transparency lines had been calculated, using a single oscillator model of dispersion correlations and the Kramers-Kronig integrals. The excitons' parameters in quantum wells (QW) and quantum dots (QD) had been determined.

Keywords: quantum wells, quantum dots, VCSEL, heterostructures.

I. Introduction

Devices based on heterostructures with QW are used in many domains, including optical communication systems. The quantum wells parameters can be determined by studying optical transitions responsible for the electronic processes inside QW and QD.

II. Experimental method

The measurement of optical spectra had been carried out using МДР-2, JASCO-670 and СДЛ-1 spectrometers at 10, 20 and 300K. The cooling of samples was made using a cryostat of closed cycle LTS-22-S-330 Workhorse-tip optical cryogenic system.

III. Experimental results and discussions

The presence of heavy and light holes in quantum wells leads to the appearance of electronic transitions series from their quantum levels. The valence bands of the light and heavy holes in $In_{0.3}Ga_{0.7}As/GaAs$ heterostructures determined the appearance of the electronic transitions hh1-e1 and lh1-e1, hh2-e2 and lh2-e2 etc. In addition, the electrons and holes are binding in excitonic states forming their ground and excited states hh1-e1 (1s, 2s, 3s...) and lh1-e1(1s, 2s, 3s...), hh2-e2 (1s, 2s, 3s...) and lh2-e2 (1s, 2s, 3s...).

Figure 1 shows transparency spectra of $In_{0.3}Ga_{0.7}As/GaAs$ heterostructure with quantum wells at 20K. Thin maxima are revealed in transparency spectra, which are conditioned by the ground (1s) and excited (2s, 3s) states of the excitonic transitions hh1-e1, lh1-e1 in the quantum well. The energetic position of the observed transitions in QW is indicated with numbers on this figure. The energy of the excitonic line in QW is determined by the correlation (1) [1, 2]:

$$E_{ex}^{l} = E_{g} + E_{0e} + E_{0h} - \frac{me^{4}}{2\mathbf{h}^{2}e^{2}(l+0.5)^{2}}$$
(1)

where *l* - quantum number, $\mathbf{\mu} = m_h^* m_e^* / (m_h^* + m_e^*)$ – excitonic binding energy.

$$R = \frac{me^4}{2\mathbf{h}^2 e^2} \,. \tag{2}$$

The excitonic binding energy (R) was determined from the energetic position of the lines lh1-e1(1s,2s,3s...) conditioned by the light holes (lh) and from the heavy holes' energies hh1-e1(1s,2s,3s...). Table 1 shows the obtained results. The binding energy R=11,5meV for the hh1-e1 excitons based on energy positions 1s-2s and based on energy lines 2s-3s the binding energy equals 12,4meV. The binding energy R=12,4meV for the lh1-e1 excitons based on the energetic position of 1s-2s lines and for those based on 2s-3s the binding energy equals 13,2meV.

Doublet structures A, B of the hh1-e1 (1s) excitons and C, D of the lh1-e1(1s) excitons are revealed in transparency spectra. The splitting value is showed on figure. The A, B and C, D splitting is conditioned by the exchange interaction of excitons. The exchange interaction of excitons, which appears in accordance with the theory of Wannier-Mott taking into account amendments to the approximation of effective mass, leads to a partial lifting of the degeneracy of the ground excitonic state and splits it to corresponding irreducible representations. The ground state of the $\Gamma_6 x \Gamma_8$ exciton is eight times degenerated in a bulk semiconductor with zinc-blend structure [1]. Spin indices of the smooth wave function take the values s = 1/2 and $m = 3/2, \pm 1/2$. The exchange interaction splits this state into three terms: $\Gamma_6 \propto \Gamma_8 = \Gamma_{12} + \Gamma_{15} + \Gamma_{25}$. In the schema of angular momentum s = 1/2 and j = 3/2 the triplet level Γ_{15} corresponds to the total angular moment j=1 with projections M = 1, 0, -1. The Γ_{12} and Γ_{25} terms correspond to the angular moment j=2 and are shifted on some value Δ_0 considering the Γ_{15} term. The splitting between Γ_{12} and Γ_{25} terms differs from zero to the extent of asphericity of the valence band Γ_8 . The splitting value caused by the exchange interaction of the singlet triplet excitonic states in bulk crystals varies from fractions of meV up to 10meV. The ground state of the 1s-exciton in the GaAs/AlAs (001) quantum well is four times degenerated. In the notation of irreducible representations of point group D_{2d} it is present $\Gamma_6 x \Gamma_6 = A_1 + A_2 + E$. Consequently, taking into account the exchange interaction this state is split into a radiation doublet E with the projections $M = s + m = \pm 1$ of the angular momentum on the Z axis and the terms A₁, A₂ (s = $\pm 1/2$, t = $\pm 3/2$) [1, 2]. The last ones are symmetrized and antisymmetrized linear combinations of states with momentum projection ± 2 . The slitting between them is small, usually neglected. The states ± 1 are dipole active at σ + and σ _ polarizations, respectively. In an anisotropic quantum well the symmetry of the system decreases and the radiative doublet should be split into two sublevels, which orientation is determined by the form of the localizing potential. Indeed, the electron-hole exchange interaction partially lifts the degeneracy of the exciton states and leads to splitting of the exciton 1s level into sublevels, corresponding to the irreducible representations. Radiation maxima conditioned by the ground (1s) and excited (2s,3s) states of the exitonic transitions e1-hh1 and e1-lh1 and, also, peaks of e1-hh2, e1- lh2 transitions are revealed in luminas and spectra at 10 K if exciting with a 542nm laser line (fig. 3). The doublet A, B and C, D is faintly observed in luminescence spectra.



Fig.1 Transparency and luminescence spectra conditioned by the ground (1s) and excited (2s,3s) states of excitonic transitions hh1-e1, lh1-e1 in the quantum well of In_{0.3}Ga_{0.7}As/GaAs heterostructure.

Figure 2 shows the transparency spectra conditioned by the ground (1s) and excited (2s, 3s) states of the excitonic transitions hh2-e2, lh2-e2 in quantum well of the $In_{0.3}Ga_{0.7}As/GaAs$ heterostructure. The measurements were done at S-P and P-P polarizations of lightwaves and 20K temperature value. The binding energy R=16,5meV for the hh2-e2 excitons based on the energetic position of 1s-2s lines. The binding exciton energy lh2-e2 basing on the energetic position of 1s-2s lines the binding energy equals 23.5meV.

Due to compression (tension) of the crystal lattice during the growth of heterostructures the lattice symmetry in strained semiconductor layers changes. In this case the valence band is shifted as a whole, the subbands of light and heavy holes are split in the center of the Brillouin zone and the effective masses of electrons and holes change. The presence of two close exciton resonances in a quantum well is taken into account by using the following dielectric function of the quantum well material:

 $\mathbf{\Sigma}_{\mathbf{i}}(\mathbf{j} = \mathbf{i})^{\mathbf{i}} \mathbf{Z}_{\mathbf{i}}(2\omega_{\mathbf{i}}LT^{\dagger}(\mathbf{j})) \omega_{\mathbf{i}} \mathbf{0}^{\dagger}(\mathbf{j}) \mathbf{j} \mathbf{1}(\omega_{\mathbf{i}}\mathbf{0}^{\dagger}(\mathbf{j})) \mathbf{1}^{\mathbf{i}} \mathbf{2} - \omega^{\mathbf{i}} \mathbf{2} + (\mathbf{k}K^{\dagger} \mathbf{2} \omega_{\mathbf{i}} \mathbf{0}^{\dagger}(\mathbf{j})) \mathbf{j} \mathbf{1}^{\dagger}(\mathbf{j}) \mathbf{j} - i\omega \mathbf{\Gamma}^{\dagger}(\mathbf{0}) \mathbf{j} \mathbf{j} \mathbf{0}$ (3) where ω – frequency, K – wave vector value, $\varepsilon_{\mathbf{b}}$ – background dielectric constant. The index of the exciton resonance j takes value j=1 for the heavy hole exciton for (h) and j=2 for the light hole exciton (l). Usually, only one resonance mode is considered [2-4]. For the excitons of j-type $\omega_0^{(j)}$ - resonant frequency, $\omega_{LT}^{(j)}$ - frequency of the longitudinal transversal splitting, $\Gamma^{(j)}$ – damping factor, $M^{(j)}$ – translational mass of exciton in a quantum well. It is significant that in case of two near exciton resonances, such as the resonances associated with light and heavy holes, the ratio between $\omega_{LT}^{(j)}$ and the splitting of the bands of light and heavy holes is extremely important. The longitudinal transversal splitting $\omega_{LT}^{(j)}$ may be more or less than the splitting of the bands of heavy and light holes. This imposes certain requirements for the analysis of optical reflection and absorption spectra of excitonic resonances of light and heavy holes.



Fig. 2 A - Transparency spectra conditioned by the ground (1s) and excited (2s, 3s) states of excitonic transitions hh2-e2, lh2-e2 in quantum well of the $In_{0,3}Ga_{0,7}As/GaAs$ heterostructure; **B** - Spectral dependence of the optical functions n, k, ε_1 and ε_2 obtained from the calculation of the reflection spectra using relations Kramers - Kronig. The inset shows the spectra of the real part of dielectric constant ε_1 for S-P and P-P polarization of the light waves in the energy region hh1-e1(lh1-e1) and hh2-e2(lh2-e2); **C** - Experimentally measured (exp.) and calculated by the dispersion relations (cal.) contours of the reflection spectra of exciton transitions hh1-e1(1s) and hh1-e1(2s) at 20K temperature.

To calculate the effective mass of holes the background dielectric constant of ε_b quantum well material in the beginning of excitonic resonance energy is necessary. The shape of reflection spectra at these temperatures was calculated to determine the background dielectric constant at 20 and 300K (fig. 3).

For the oscillator with the resonance frequency $\omega_1 = 0.98 \text{eV}$ at 20K $\Delta \omega_{LT} = 69 \text{meV}$, $\Gamma = 18 \text{meV}$ and $\varepsilon_B = 7$, and for the oscillator with resonance frequency $\omega_2 = 1.086 \text{eV}$ $\Delta \omega_{LT} = 75 \text{meV}$, $\Gamma = 7.8 \text{meV}$ and $\varepsilon_B = 7$. The change of the value of effective mass M has a little effect on the shapes of the calculated curves. At 300K the best agreement between experimental and calculated shapes are obtained with the following parameters: for $\omega_1 = 0.905 \text{eV}$ $\Delta \omega_{LT} = 70 \text{meV}$, $\Gamma = 12.8 \text{meV}$ and $\varepsilon_B = 6$, for $\omega_2 = 1.016 \text{eV}$ $\Delta \omega_{LT} = 70 \text{meV}$, $\Gamma = 15.8 \text{meV}$ and $\varepsilon_B = 5.5$, and for $\omega_3 = 1.12 \text{eV}$ $\Delta \omega_{LT} = 55 \text{meV}$, $\Gamma = 27 \text{meV}$ and $\varepsilon_B = 4.6$. The obtained values $\Delta \omega_{LT} = 70 \text{meV}$ characterize two intensive minimum in the reflection spectra, which are due to QD. The value of $\Delta \omega_{LT}$ is overstated due to the fact that in the calculation model the transverse mode frequency corresponds to the maximum R at $\omega_1 = 0.905 \text{eV}$, and the second oscillator ω_2 =1.016eV. At the same time the real value of the transverse frequencies of the oscillators correspond to the minima of the reflection spectra located at higher energies This method is used in calculations of exciton polaritons spectra in bulk crystals [5], which we used only to determine the background dielectric constant value ε_B in the beginning of the electronic transitions region in QW. The right part of figure 3 shows the experimental and calculated reflection spectra (1-R) in the model of excitonic polaritons in QW [1, 2].

Calculation of reflection spectra of In_{0.3}Ga_{0.7}As/GaAs heterostructure using the Kramers -Kronig relations were made to determine the optical constants. Figure 2, B shows the spectral dependence of optical constants n, k, ε_1 and ε_2 at room temperature in a wide range of energies. The data obtained shows that the most strongly changes are the dielectric function ε_1 and ε_2 and resonant frequency value of QD. Measurements and calculations were made in S-P P-P polarized light waves to estimate the magnitude of the resonances ε_b QW, i.e. the transition energy region hh1-e1(lh1-e1) and hh2-e2(lh2-e2). In S-P polarization the reflection coefficient R is greater than in P-P polarization (in the transmission coefficient is found an inverse relationship). In S-P polarization the hh2e2(1s, 2s) transitions occur and therefore $\varepsilon_b(\varepsilon_1)=6$ describes these transitions. The dielectric constant $\varepsilon_{\rm b}(\varepsilon_1)=6.9$ for this polarization in the energy range 1.16eV, i.e. hhl-el transitions. In case of the energies 1.16 eV and P-P polarization $\varepsilon_b(\varepsilon_1) = 5,5$ and of 1.3 eV the energy $\varepsilon_b(\varepsilon_1) = 4,3$. As it can be seen the value of the background dielectric constant obtained from the calculation of the reflection spectra using the Kramers-Kronig relations are almost consistent with the calculations of the dispersion correlation. Thus, in the region of the exciton transitions hh1 - e1 (lh1 - e1) the difference between the dielectric constant $\Delta \epsilon = \epsilon_b^{hh} - \epsilon_b^{lh} = 6.0-4.3=1.7$ and region of hh2 - e2 (lh2 - e2) transitions $\Delta \varepsilon = \varepsilon_b^{hh} - \varepsilon_b^{hh} = 6.9 - 5.5 = 1.4$. As it can be seen within the experimental error, these values are the same. The effective mass of electrons $m_e^* = 0.05m_0$ was used to estimate the effective mass of the heavy and light hole and the binding energies of (R) defined above were used to calculat mass of the excitons m. The exciton's binding R=0.0124eV, the effective mass m=0.0447m₀. The effective mass $m_{hh}^{*}=0.32m_{0}$ for the background dielectric constant $e_{B}=6.9$. The exciton binding energy R=0.0132eV, effective mass m=0,0293m₀ for transitions lh1-e1. The effective mass $m_{lh}^*=0.07m_0$ for the background dielectric constant e_{B} =5.5. The obtained values of effective masses of light and heavy holes are consistent with those obtained for the heterostructures in [6, 7].



Fig.3 Experimentally measured (exp.) and calculated by the dispersion relations (cal.) contours of the reflection spectra at 20K and 300K temperature

The reflection spectra measured at 20K and their contours are calculated by dispersion relations in the region of exciton transitions hh1-e1(1s) and hh1-e1(2s) on the structures of which the transmission spectra presented in figure 2, C were obtained. The best agreement between experimental and calculated data was obtained for the oscillator $\omega_0 = 1.1629$ eV, with the following parameters: $\Delta\omega_{LT} = 1.1$ meV, $\Gamma=0.3$ meV and $\epsilon_B=6.7$. Figure 3 shows also the scheme of optical transitions in the ground and excited states of excitons of QW (right panel of the figure). Longitudinal transverse splitting $\Delta\omega_{LT}$ from the calculation of reflection spectra hh1-e1 is greater than for the shape hh1-e1(1s). This is because the contour of the reflection shape hh1-e1 contains the total contribution of both ground and excited excitonic states of heavy and light holes.

The exciton binding energy R=0.0165eV, effective mass m=0.0436m₀ for hh2-e2 transitions. The effective mass $m_{hh}^*=0.30m_0$ for the background dielectric constant $e_B=6.0$. The exciton binding energy R=0.022eV, effective mass m=0.0293m₀ for lh2-e2 transitions. The effective mass $m_{lh}^*=0.07m_0$ for the background dielectric constant $e_B=4.5$. The parameter M (M=m_{hh}^*+m_e^*), obtained from the calculation of the reflection spectra shapeof the oscillator and of exciton transition hh1-e1(1s) is equal to 0.4m₀. Consequently, the effective mass $m_{hh}^*=M-m_e^*=0.4-0.07=0.33m_0$.

IV. Conclusions

In the reflection, transmission and luminescence spectra the lines caused by the transitions hh, lh1-e1 (1s, 2s, 3s), hh2, lh2-e2 (1s, 2s, 3s), hh1, lh1-e2 (1s), and hh3, lh3, -e3 (1s) have been detected. Calculations of the reflection and transmission contour lines in single-oscillatoric model of dispersion relations and the Kramers-Kronig integrals show that the values of background dielectric constant in the immediate vicinity (the beginning) of transitions in the ground and excited states of excitons in QW differ. The energy of the excitons hh-e1, lh1-e1, effective masses m_{hh}^* and m_{lh}^* , and the damping factor for optical transitions in QW and QD were determined. The value of the damping factors for QD₁-QD₄ and exciton levels of QW do not differ very much. The radiative lifetime of excitons in quantum dots and excitons in quantum well in the considered structure varies in the limits of $\tau_0 = (2\Gamma_0)-1 \cong 0.04 - 0.1$ ps.

V. References

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