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Effect of additional illumination on the kinetics of exciton complex formation in the quantum wells of undoped GaAs/AlGaAs structures

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Abstract. Low-temperature \((T = 2 \text{ K})\) photoluminescence (PL) and photoluminescence excitation (PLE) spectra of GaAs/AlGaAs \((x = 0.05)\) structures with shallow quantum wells (QW) and the effect of additional illumination by He–Ne laser radiation on these spectra were investigated. It was found that the PLE spectra exhibit a number of broad bands in the above-barrier energy region; these bands alternate “in opposite phases” in the spectra of free excitons and excitonic complexes (trions) (i.e., an increase in the exciton luminescence intensity is accompanied by a decrease in the luminescence intensity of the complexes). In the case where the photon energy of the Ti-sapphire laser is tuned to excite the QW states, additional illumination results in the shift of the equilibrium in the exciton–trion system towards an increase in the concentration of the latter species. On the other hand, upon excitation into certain barrier states with energy both below and above the barrier gap, additional illumination shifts the equilibrium in the opposite direction.

Charged three-particle excitonic complexes (trions) in low-dimensional semiconductor structures were actively investigated in the recent years. In particular, it was demonstrated that, in GaAs/AlGaAs structures without intentional doping, negatively or positively charged localized trions can be formed due to the preferential capture of electrons or holes into the quantum wells (QWs), as well as due to the presence of residual impurities in the barrier layers [1–4].

It is of substantial interest to follow the variation of the “molecular composition” of the excitonic system as the photon energy of the exciting radiation is continuously tuned in the range including the band gap of the barrier layers, i.e., under the transition from the excitation into the QWs to the above-barrier excitation. This can be carried out for GaAs/AlGaAs structures with low Al content in the barrier layers.

The structure under study contains two shallow tunneling-isolated GaAs QWs of width 4 and 3 nm, separated by a 60-nm-wide Al\(_{0.05}\)Ga\(_{0.95}\)As barrier and confined from both sides by 100-nm Al\(_{0.05}\)Ga\(_{0.95}\)As layers. The band gap in the barrier layers \(E_{\text{gb}} \approx 1.6 \text{ eV}\), and the tuning range of a Ti-sapphire laser (in our case, 1.49–1.77 eV) is sufficient to perform the measurements discussed above. In addition, these structures have a relatively simple energy spectrum (there is just one single-particle quantum-confinement level for each kind of quasiparticles in each QW [5]), which makes data analysis and interpretation more straightforward.

The photoluminescence (PL) spectrum of the narrow QW of the structure under the He–Ne laser excitation (photon energy 1.959 eV) is shown in Fig. 2 (inset, upper curve). The spectrum consists of two lines, the higher-energy one being related to the recombination radiation of the free heavy-hole excitons (FE) and the lower-energy one, to the recombination radiation of the excitonic complexes (EC). The EC and FE PL excitation (PLE) spectra for this QW (recorded with the monochromator output slit fixed at the corresponding PL lines) are shown by solid lines in Figs. 1 and 2, respectively. The shape of the PLE spectra
of the free excitons and the complexes differ considerably when the photon energy exceeds the barrier band gap $E_{gb}$: there are broad bands whose position do not coincide in the FE and EC spectra; moreover, the maxima and minima in both curves alternate “in opposite phases”, i.e., an increase in the excitonic complex PL intensity is accompanied by a decrease in the PL intensity of the free excitons. Thus, the shape of the QW PL spectrum depends significantly on the photon energy of exciting radiation.

It seems likely that the variation of the luminescence intensity related to the free excitons and excitonic complexes with the energy of exciting photons in the above-barrier spectral region is due to the dependence of the efficiency of the photoexcited charge carrier capture into the QW on their initial energy. The concentration $n_T$ of the excitonic complexes depends on the density $n$ of the electron (or hole) gas and the exciton concentration $n_{ex}$ in the QW ($n_T \sim n_{ex} \cdot n$). Therefore, the efficiency of the excitonic complex formation should increase when the nonequilibrium charge carriers of one sign are preferentially captured into the QW. The energy width of the bands in the EC PLE spectrum (Fig. 1) gives an indication of the charge carrier energy region where such a situation does take place. Under these conditions, the density of the exciton gas in the QW should decrease due to exciton binding into trions.

The density of electron (hole) gas and even the type of the conduction in the QW can be changed by an additional illumination of the sample by the radiation with the photon energy exceeding the band gap in the barrier layers [2, 3]. The effect of the additional illumination of the sample by the He–Ne laser radiation on the PLE spectra of the EC and FE in the narrow QW is shown in Figs. 1 and 2, respectively. When the QW states are excited (the Ti-sapphire laser photon energy $h\nu_{exc} < 1.592$ eV), additional illumination results in an increase of the EC luminescence intensity (Fig. 1), which is accompanied by a decrease
Fig. 2. Excitation spectra of the free exciton PL without and with additional illumination by a He–Ne laser. Inset shows the PL spectra: upper part, luminescence excited by a He–Ne laser; lower part, differential spectra (see the text) for two excitation photon energies.

of the intensity of the FE luminescence (Fig. 2). A more complicated picture is observed when we excite the above-barrier states or the states with the energies slightly below but close to $E_{\text{ub}}$ responsible for the triplet in the PLE spectra at $h\nu_{\text{exc}} \approx 1.592-1.599$ eV. While the additional illumination leads to an increase in the intensity of the free-exciton PL in the whole range of excitation photon energies $h\nu_{\text{exc}} > 1.592$ eV, the intensity of the PL of excitonic complexes can either increase or decrease. Moreover, at certain values of the excitation photon energy (e.g., $h\nu_{\text{exc}} \approx 1.595, 1.636$ eV etc.) the phase of the photodetector signal changes in such a way that negative signal appears at the output of the lock-in amplifier (Fig. 1). Note that the energy difference between the values of $h\nu_{\text{exc}}$ corresponding to the regions of negative signal in the PLE spectra under additional illumination (Fig. 1) is of the order of the optical phonon energy in the barrier layers.

The influence of additional illumination on the radiative recombination processes in the QW, seen in the PLE spectra, is also reflected very clearly in the PL spectra. The PL spectra obtained, like the PLE spectra in Figs. 1 and 2, under the excitation by modulated Ti-sapphire laser radiation and the additional stationary illumination by a He–Ne laser are shown in the inset to Fig. 1. In the case of resonance excitation into the QW light-hole exciton states ($h\nu_{\text{exc}} = 1.582$ eV), He–Ne illumination results in enhanced EC luminescence intensity because of an increase in the trion concentration, which takes place due to the binding of the excitons created by the excitation radiation with the nonequilibrium charge carriers in the QW originating from the additional illumination. In the case of excitation into the states corresponding to the low-energy component of the triplet ($h\nu_{\text{exc}} = 1.595$ eV), charge carriers of opposite signs are created in the QW by the excitation and the additional-illumination radiation, which leads to a decrease in the stationary concentration of the charge carriers and, thus, of the trions. This brings about the appearance of the negative
signal in the PL and PLE spectra (Fig. 1). Such a behavior of the electron–hole system is also reflected in a something different way in the differential (with respect to the additional illumination intensity) PL spectra recorded under the conditions of stationary excitation by a Ti-sapphire laser and modulated additional illumination (Fig. 2, inset). In this case, the changes in the PL spectra related to the additional illumination are recorded.

The PL and PLE spectra of the wide QW of the structure exhibit similar behavior to that of the narrow QW.

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References