LUMINESCENT PROPERTIES OF NEAR-SURFACE SEMICONDICTIVE LAYERS AND QUANTUM SUPERLATTICES

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The new results of studies of the GaAs/AlAs quantum superlattices (QSLs) of the I and II types and the interfaces of heterostructures (surface effects) using the pulse (femtosecond) light excitation technique have been presented. The peculiarities of the photoluminescence relaxation of QSLs with various thicknesses of Q-layers (GaAs) and barriers (AlAs) are analyzed. It is demonstrated that, at a high excitation level, the electron-hole plasma appears in the quasidirect-gap QSLs, where the density of free carriers is by more than one order larger than that possible for the bulk of GaAs. By studying the spectra of spontaneous and stimulated emissions, we calculate the optical gain coefficient as a function of the pumping density for nonlinear effects.

1. Introduction

The development of modern integrated electronics and optoelectronics stimulates wide studies of two-dimensional and quasi-two-dimensional effects of various types running in spatial bounded layered systems: on the surface of semiconductors, on the interfaces of heterostructures, and in quantum superlattices (SLs) [1–7]. In this case, the application of the highly sensitive nondestructive method of photoluminescence (PL) to the study of fundamental characteristics of the mentioned system is of significant importance. Basic is the question about the formation and the manifestation of the nonequilibrium electron-hole plasma (EHP) on the interfaces of layered "dielectric–semoconductor" structures [8, 9] and in layered semiconductors [10, 11] in the optical spectra. A number of peculiarities of nonlinear optical effects related to a high concentration of nonequilibrium charge carriers should be expected for a two-dimensional EHP as compared with a three-dimensional one under identical conditions of their excitation, in particular, those in the processes of stimulated emission in the surface regions of semiconductors and in quantum-dimensional structures with superthin layers (quantum superlattices of the I and II types). In the present work, the results of luminescence studies of GaAs/AlAs quantum SLs of the I and II types and the interfaces of GaAs/AlGaAs heterostructures with the use of pulse (femtosecond) excitation sources are given. In particular, we have analyzed the specific features of the PL kinetics of GaAs/AlAs SLs with various widths of quantum wells (GaAs) and barriers (AlAs). The optical gain spectra in GaAs/AlGaAs quantum superlattices are experimentally measured and theoretically calculated.

2. Decay of Photoluminescence in I-type Superlattices

The investigation of the PL decay kinetics of quantum superlattices gives an important information about their energy spectra. In particular, challending is the time interval $\Delta t = 10^{-9}$–10^{-12} s after the excitation of a specimen by a laser pulse, during which the change of mechanisms of recombination (participation of light and heavy holes, free and bound excitons, *etc.*) occurs.

The study of the PL decay with the use of pulse lasers with a duration of pulses $\sim 10^{-13}$ s allows one to establish the types of radiative electron-hole transitions, time characteristics of various mechanisms of recombination, resolution of peaks which are close in energy and differ by recombination times [12].

The typical spectrum of a direct-gap superlattice is composed from a single PL band caused by the recombination of heavy excitons (electron – heavy hole) localized

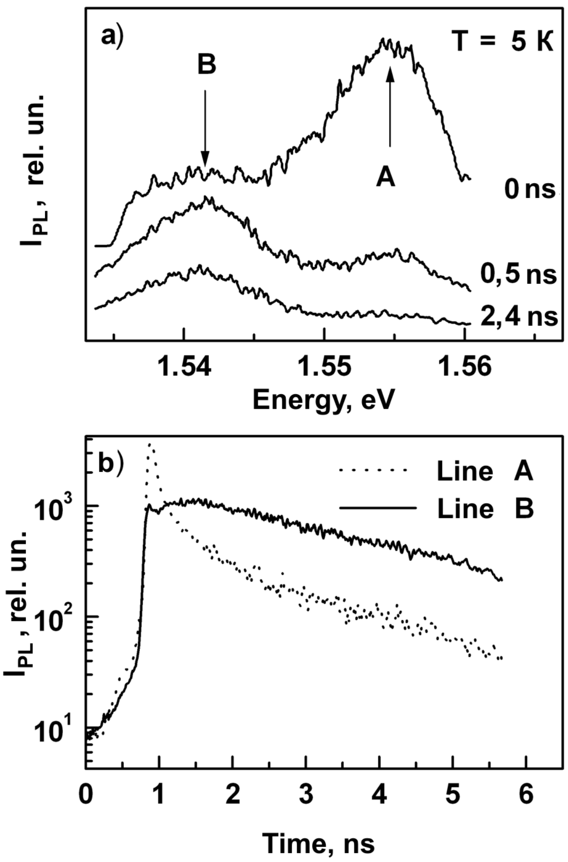

Fig. 1. (*a*) PL spectra of a 36/36 SL at $T = 5$ K obtained at various time delays after the exciting pulse; (*b*) the time behavior of the PL intensities of lines A and B

in GaAs layers. However, some studies revealed the additional PL band, which was assigned to light excitons composed of electrons and light holes.

In Fig. 1, we show the PL spectra of a GaAs/AlAs 36/36 SL (36/36 are the numbers of monolayers GaAs and AlAs, respectively) obtained at various time moments after the exciting laser pulse. The PL spectrum includes two lines denoted in Fig. 1,*a*, respectively, by A and B, whose maximum energies are 1.555 and 1.541 eV. The difference of the energies of 14 meV is close to the calculated splitting of the subbands of heavy and light holes (18 meV). However, these calculations do not involve the difference of the binding energies of heavy and light excitons, which is equal to ~ 2 meV for the given thicknesses of the wells.

The kinetics of lines A and B are different (see Fig. 1,*b*). Whereas line A demonstrates the bi-exponential decay with the time constants ~ 100 ps and ~ 1.7 ns at 5 K, the intensity of line B increases firstly with the time constant ~ 230 ps and then decreases with the time constant ~ 2.7 ns. The initial decay of line A and the simultaneous increase in the intensity of line B can be explain by the transition from light to heavy excitons.

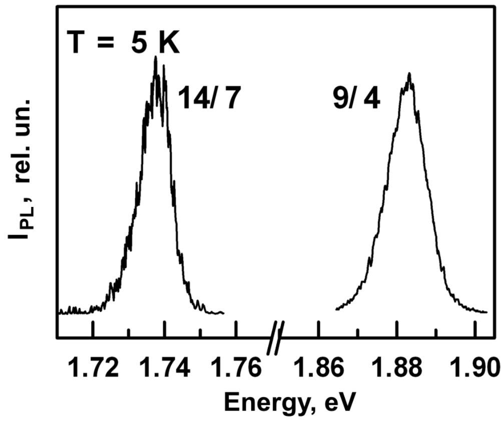

Fig. 2. PL spectra of 14/7 and 9/4 SLs at $T = 5$ K. Spectra are normed to their maxima

The long-duration components correspond to the lifetimes of excitons of two types. Their growth with the temperature allows us to conclude that the observed excitons are free.

Then we consider the PL kinetics of specimens with less thicknesses of quantum layers (14/7 and 9/4 SLs). Both SLs also belong to direct-gap SLs of the I type. The PL spectra of these SLs given in Fig. 2 are presented by a single narrow line caused by the recombination of excitons (electron - heavy hole) localized in layers of gallium arsenide.

We note that the width of the quantum well of a 9/4 SL is less than the critical one, at which the transition to quasidirect-gap SLs of the II type (12 monolayers) occurs. However, the barrier width less than the well width makes the given SL to be direct-gap with the conduction band bottom localized at the Γ-point of GaAs.

The studies of the time-resolved PL spectra of a 9/4 SL show that, in the whole temperature interval (5–100 K), the decay of the PL maximum is described by a single exponent (Fig. 3), which testifies to the localization of excitons even at rather high temperatures. It is worth noting the quite strong dependence of the PL decay duration for a 9/4 SL on the temperature, as distinct from the case of the specimens with wider wells: at low temperatures ($T > 30$ K), the decay duration is practically invariable, and the increase of the temperature from 30 K to 80 K leads to a decrease of the decay duration from ~ 200 ps down to ~ 30 ps.

The results obtained can be summarized as follows. In direct-gap SLs where the well width is comparable with the exciton radius (like that in a 36/36 SL), the PL spec-

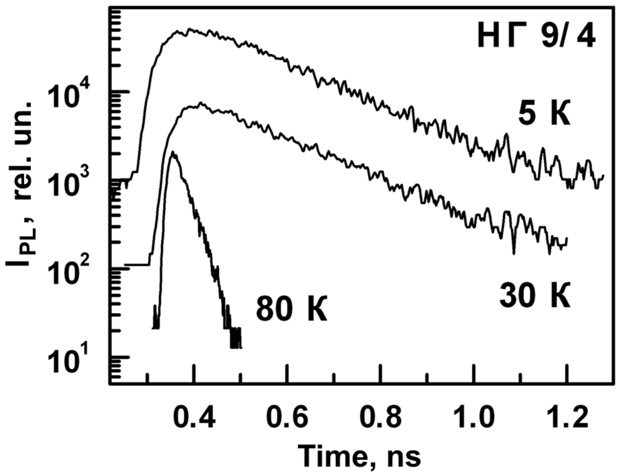

Fig. 3. Time variations of the PL maximum intensity of a 9/4 SL at $T = 5$, 30, and 80 K

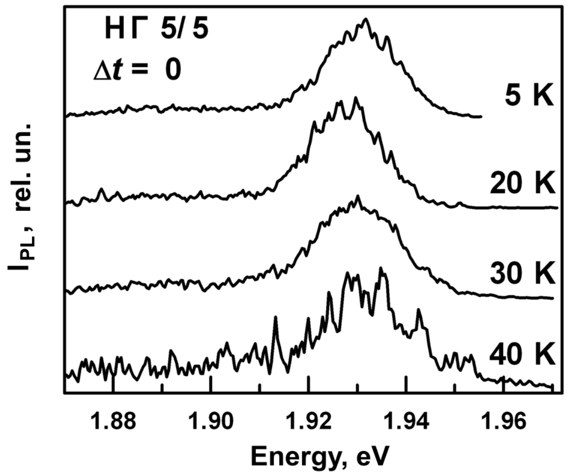

Fig. 4. TRPL spectra of a 5/5 SL at the zero delay after the exciting pulse ($\Delta t = 0$) at various temperatures

tra contain two bands of free excitons, which are formed with the participation of heavy and light holes. In SLs where the well width is less by a factor of ~ 1.5 than the radius of an exciton, which turns out to be "clamped" by the well walls, the free excitons localize theselves rapidly (for ~ 150 ps) on the heteroboundaries of GaAs-AlAs. In a 14/7 SL with thinner layers of quantum wells (~ 0.4 exciton radius), the emission at low temperatures is caused by bound excitons, whose partial delocalization starts only at the increase of the temperature ($T \geq 80$ K). In direct-gap GaAs/AlAs SLs with superthin layers (SL 9/4), the emission of localized excitons is observed in the entire interval of temperatures under study.

3. Kinetics of Photoluminescence of Short-Period GaAs/AlAs Superlattices of the II Type

Here, we describe the results of studies of the time-resolved photoluminescence (TRPL) spectra of two GaAs/AlAs SLs of the II type. One of the specimens, a 5/5 SL, belongs to quasidirect-gap SLs, whose PL is caused by the recombination of spatially separated excitons composed of X_z electrons of the barrier and heavy holes localized in a GaAs layer.

In Fig. 4, we present the TRPL spectra of a 5/5 SL, which are obtained at various temperatures and at the zero time delay after the excitation. In the temperature interval $T = 5$–20 K, we observe a shift of the PL maximum to the long-wave side by ~ 3 meV after a time delay, whose value depends on the temperature and varies from ~ 1 μs at 5 K to ~ 100 ns at 20 K.

The shown specific features of the PL spectrum of a 5/5 SL are characteristic of the inter-impurity (donor-acceptor) recombination. In this case, the energy $E_{\rm ph}$ of an emitted photon depends on the distance between the electron and the hole by the law

$$E_{\rm ph} = E_g - (E_{\rm D} + E_{\rm A}) + \frac{e^2}{\varepsilon r_n}, \qquad (1)$$

where E_g is the bandgap width, $E_{\rm D}$ and $E_{\rm A}$ are the ionization energies of, respectively, the donor and acceptor centers, r_n is the distance between the donor and the acceptor, and ε is the dielectric constant.

Indeed, a shift of the PL band to the long-wave side during the decay means that the pairs with close bound electrons and holes, which contribute to the high-energy part of the spectrum, recombine significantly faster than the pairs with separated electrons and holes, which tunnel through a larger distance prior to the recombination and contribute to the low-energy region of the spectrum. In our case, the localization centers are fluctuations induced by the inhomogeneity of heteroboundaries. In this case, the closely located electrons and holes create, due to the Coulomb interaction, specific localized excitons. A decrease of the recombination time of such excitons with increase in the temperature is caused by the delocalization of charge carriers and, respectively, by the faster recombination (without tunneling).

On the other hand, the indicated peculiarity can be explained by the spectral migration of localized excitons by means of, obviously, the emission of acoustic phonons from more shallow to deeper localization centers. The shift of the PL maximum energy with increase in the temperature corresponds to the temperature dependence

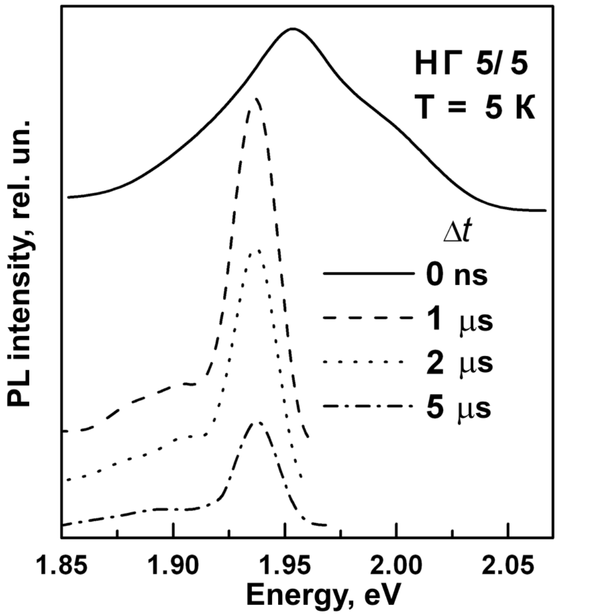

Fig. 5. PL spectra of a 5/5 SL obtained at various time delays for high levels of excitation

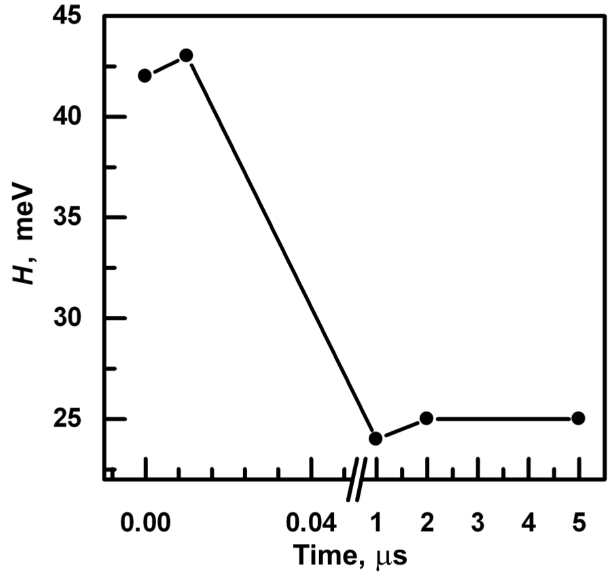

Fig. 6. Time dependences of the half-width H of the PL zero-phonon line of a 5/5 SL at high levels of excitation. $T = 5$ K

of the bandgap widths of GaAs and AlAs, taking into account that they vary identically [13]. At $T = 30$ K, the inverse situation is observed (Fig. 4): the spectral maximum energy at the zero delay is greater by ~ 2 meV than the corresponding value at 20 K. In the course of time like the previous cases, we also observe a red shift of the maximum. However, it is as high as ~ 6–7 meV in this case, which is twice more than that at lower temperatures. This testify that the excitons are free at the initial time moment after the excitation and localized with time on the inhomogeneities of heteroboundaries. The value of red shift corresponds approximately to the localization energy of excitons. It is characteristic that, at $T = 5$–30 K, the shape of PL decay curves depends on the detection energy and varies from a nonexponential shape on the short-wave end of the spectrum to the exponential one on the long-wave end.

We also studied the PL kinetics of a 5/5 SL at high levels of excitation with the use of a pulse nitrogen laser ($P \sim 3$ kW, the pulse half-width of 10 ns). The PL spectra obtained under such conditions are shown in Fig. 5.

We pay attention to the appearance of a wide band of PL ($E_{\max} = 1.951$ eV) with the half-width $H \cong 43$ meV at once after the excitation. During ~ 10 ns, this band shrinks to $H \cong 15$ meV and shifts to the long-wave side by ~ 20 meV. Then its energy position and half-width remain invariable for the whole decay duration. The time dependences of the half-width of the PL zero-phonon line are presented in Fig. 6.

The indicated peculiarities of PL at high levels of excitation in a 5/5 SL testify to the following character of recombination processes. The nonequilibrium electrons and holes excited by a laser pulse termalize in the appropriate bands and create free excitons. However, the high density of excitons leads to the formation of a nonequilibrium electron-hole plasma in the SL, whose recombination causes the appearance of the short-wave PL band. In the process of recombination, the concentration of carriers in the plasma decreases, which is confirmed by the narrowing of the PL band, and the plasma disappears approximately in 30 ns.

The nonequilibrium electrons and holes, which have no time to recombine, create localized $X_z - \Gamma$ excitons. They recombine during ~ 5 μs, which causes the appearance of the long-time component in the PL kinetics of the specimen under study.

The value of half-width of the plasma band allows us to determine the concentration of free carriers in the two-dimensional electron-hole plasma created by the powerful laser radiation:

$$2H \cong E_{\mathrm{F}_n} + E_{\mathrm{F}_p} = \frac{1}{2}(3\pi^2)^{2/3} k_0 T \hbar^2 \left(\frac{1}{m_e} + \frac{1}{m_p}\right) n^{2/3}, \quad (2)$$

where E_{F_n} and E_{F_p} are the Fermi quasilevels for electrons and holes, respectively, T is the temperature, $m_{e,p}$ are

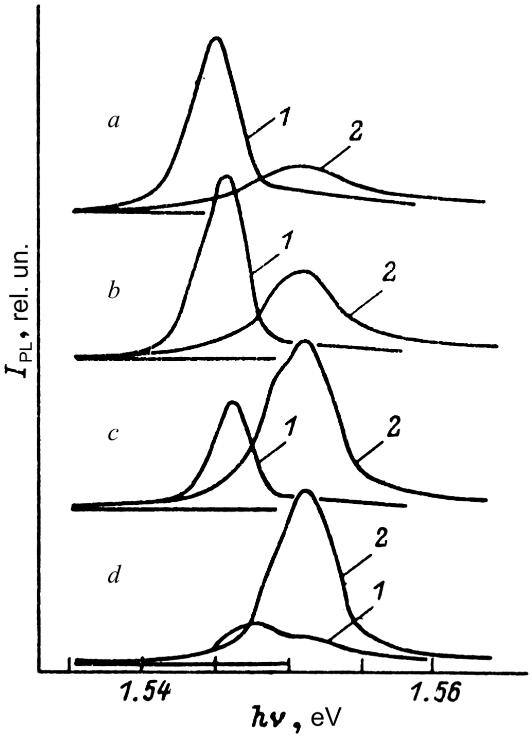

Fig. 7. Spectra of stimulated (*1*) and spontaneous (*2*) emissions of the GaAs/$Al_{0.35}Ga_{0.65}$As SL at various excitation power densities (L_0 = 4 MW/cm^2). $a - L_0$, $b - 0.2L_0$, $c - 0.05L_0$, $d - 0.02L_0$

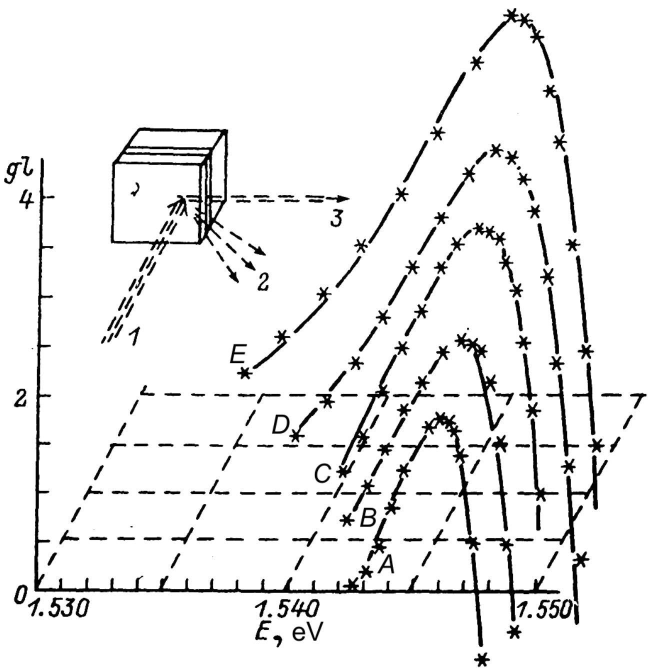

Fig. 8. Optical gain spectra of the GaAs/$Al_{0.35}Ga_{0.65}$As SL at various exciting emission densities L. Points – experiment (T = 4.2 K, l = 100 μm), continuous curves – calculations. L (MW/cm^2): a – 0.2, b – 0.44, c – 0.76, d – 1.68, e – 4. The insert shows the geometry of the experiment. *1* – laser beam, *2* – spontaneous emission, *3* – stimulated emission

the effective masses of electrons and holes, and n is the concentration of carriers in the plasma.

The obtained value $n \sim 10^{18}$ cm^{-3} exceeds the corresponding concentration of the plasma in bulk GaAs (3×10^{16} cm^{-3}) by more than one order.

4. Optical Gain in Quantum Superlattices of GaAs/Al_xGa_{1-x}As

We have studied the optical gain spectra in GaAs/Al_xGa_{1-x}As superlattices as functions of the level of the optical pumping L.

The spectra of the coefficient of optical gain were determined by the method described in [14]. The photoluminescence was excited by the second harmonic of a LTIPCh-4 laser with the active element YAG:Nd^{3+}. The excitation power density varied in the limits 0.08–4.0 MW/cm^2.

In Fig. 7, we give the spectra of stimulated (1) and spontaneous (2) PL as functions of the level of excitation. As a characteristic feature, we mention the rather low threshold of the optical pumping $L_{\min} < 0.08$ MW/cm^2, at which the stimulated emission arises. In addition, as is seen from Fig. 7, the maximum of the stimulated emission is shifted to the long-wave side as compared with the maximum of the spontaneous emission.

The spectra of the coefficient of optical gain were calculated from the ratio of the intensities of the stimulated and spontaneous emissions for the relevant wavelengths λ obtained at the same levels of excitation:

$$\frac{I_{\text{stim}}}{I_{\text{spont}}} = \frac{\exp(gl) - 1}{gl}, \qquad (3)$$

where g is the coefficient of optical gain, l is the strip excitation length (under conditions of our experiment, l = 100 μm).

The spectra of the coefficient of optical gain calculated in such a way for five values of pumping density are presented in Fig. 8.

The calculation of the optical gain spectra for the studied structures was carried out with regard for the following assumptions:

a) The executed estimates of the energy gap between the lowest levels of the electron subsystem for the given type of an SL with the widths of quantum wells $\sim$100 Å and the widths of barriers $\sim$150 Å gave the value $\sim$100 meV, which is essentially (by 6–10 times) greater than the width of the experimentally determined optical-gain bands even under the maximally high levels of excitation. Therefore, while determining the Fermi quasilevel in the electron subsystem, we can restrict ourselves by the consideration of only the lowest electron subsystem and neglect the subband of light holes under specific conditions of the experiment.

b) At high levels of excitation, the short-wave expansion of the optical gain spectrum becomes weak, as seen from Fig. 7 (as distinct from the clearly pronounced long-wave spreading of the optical gain spectrum related to the renormalization of the bandgap due to many-particle effects), which testifies to the expansion of the electron-hole plasma [15].

c) The base model of the structure under study is the model of direct interband transitions with the conservation of the quasimomentum and with the establishment of some effective temperature in the electron-hole plasma due to the low values of scattering times of charge carriers by one another.

d) The shapes of the optical gain spectra, whose characteristic feature is the elongated long-wave edge despite the two-dimensionality of the emitting electron-hole plasma, testify to the spreading of final states of recombining electrons and holes. Therefore, the fitting of the calculated and experimental optical-gain spectra was carried out under the assumption of the decay of electron-hole states, which quadratically diminishes to the Fermi energy: $\Gamma = \Gamma_0(1 - k/k_\mathrm{F})^2$ [16, 17]. Due to the spreading of states with energy E', the probability of the emission of photons with energy E was approximated by the normed Lorentzian $D(E, E') = (2\pi)^{-1}\Gamma(E')/[(E - E')^2 + \Gamma^2(E')/4]$, which is transformed at $E' > E_\mathrm{F}$ in the δ-function. The final optical gain spectrum $g(E)$ was calculated by using the convolution of the unspread gain spectrum $g'(E)$ with the Lorentzian $D(E, E')$ [18, 19],

$$g(E) = \int\limits_{E_g}^{\infty} g'(E')D(E, E')dE', \tag{4}$$

where E_g is the width of the renormalized bandgap, $g'(E) = A(f_e + f_h - 1)$, A is a constant, which is practically independent of the energy, and f_e and f_h are the electron and hole occupancy functions.

A specific feature of our calculations is the consideration of the expansion of the electron-hole plasma in the plane of quantum wells with the drift velocity V_D due to the action of forces caused by the Fermi pressure gradient on charge carriers. We make it by using the shifted

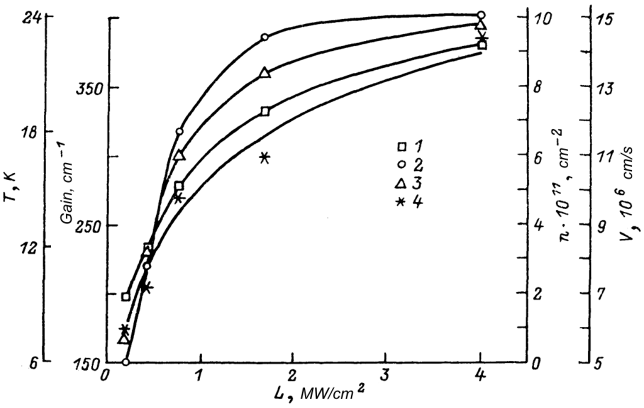

Fig. 9. Dependence of the fitting parameters on the excitation power density L: the density of electron-hole pairs n (*1*), temperature T (*2*), drift velocity V_D (*3*), and the coefficient of optical gain g (*4*), which are used at the fitting of experimental and theoretical curves $gl(E)$ (Fig. 8)

distribution functions

$$f_{e,h} = \frac{1}{\exp\left[\left(\frac{\hbar^2(\mathbf{k}+\mathbf{k}_{D_{e,h}})^2}{2m_{e,h}} - F_{e,h}\right)/k_\mathrm{B}T\right] + 1} \tag{5}$$

and averaging over the angles between $\mathbf{k}$ and $\mathbf{k}_{D_{e,h}}$, where $\mathbf{k}_{D_{e,h}} = m_{e,h}\mathbf{v}_\mathrm{D}/\hbar$, and $F_{e,h}$ are Fermi quasilevels, which are reckoned from the bottoms of relevant bands. Since we consider the direct transitions, the connection of the energy and the quasimomentum takes the form $E = \hbar^2/k^2/2\mu + E'_g$, $\mu = m_e m_h/(m_e + m_h)$, where m_h is the mass of a heavy hole. Under the given specific conditions, the Fermi quasilevels $F_{e,h}$ can be determined by the formula $F_{e,h} = k_\mathrm{B}T\ln(\exp[\pi\hbar^2 n/m_{e,h}k_\mathrm{B}T] - 1)$, where n is the concentration of charge carriers, and T is the effective temperature of the electron-hole subsystem.

As a result of calculations of the optical gain spectra by the above-presented formulas at $\Gamma_0 = 1$ meV and their fitting to those obtained experimentally (Fig. 8), we determined the parameters of the electron-hole plasma T, g, n, and V_D at the corresponding levels of excitation (Fig. 9).

Thus, the intense laser radiation causes the formation of the two-dimensional electron-hole plasma in a quantum GaAs/AlGa$_{1-x}$As SL. The analysis of the shape of the optical gain spectra with regard for the effect of expansion allowed us to find the following parameters of the electron-hole plasma: the concentration of nonequilibrium carriers, their overheating, and the drift velocity of charge carriers, which appears due to the Fermi pressure. The value of drift velocity ($V_\mathrm{D} = 10^7$ cm/s) approaches the limiting velocity of transfer of electrons and holes in solids. The coefficient of optical gain for the SL under study is greater by a factor of 1.3 than that for a GaAs/AlGa$_{1-x}$As heterostructure at the same levels of excitation.

5. Conclusions

The studies of time-resolved PL spectra of GaAs/AlAs SLs of different types (direct-gap, quasidirect-gap, indirect-gap ones) in a wide interval of temperatures allowed us to establish some peculiarities of the recombination processes running in such structures.

In particular, we observed free heavy and light excitons and the "light-heavy excitons" transition in direct-gap SLs with the thickness of wells of the order of the exciton radius at once after the excitation. For a I-type SL with the thickness of GaAs layers less than the exciton radius at low temperatures ($T < 70$ K), the free excitons are localized during ~ 150 ps on the inhomogeneities of heteroboundaries and then emit, by forming a new PL band. In an SL with a more less thickness of quantum layers, the emission at low temperatures is caused by bound excitons, whose partial delocalization starts only at the increase of the temperature ($T \geq 80$ K). Finally, we observed the emission of localized excitons in direct-gap GaAs/AlAs SLs with superthin layers (9/4 SL) in the whole studied interval of temperatures.

A decrease in the well width significantly accelerates the decay of PL, which can be explained by an increase in nonradiative losses.

SLs of the II type ($n = m < 12$ monolayers) are characterized by a significant influence of fluctuations of the thickness of quantum layers on the relaxation of charge carriers, which increases with decrease in the thickness of layers and causes a considerable decrease in the radiative lifetime of excitons. At high levels of excitation, the electron-hole plasma is formed in quasidirect-gap specimens. In the plasma, the concentration of carriers exceeds the corresponding value for bulk GaAs crystals by more than one order.

The study of the spectra of spontaneous and stimulated emissions of the electron-hole plasma in GaAs/Al$_x$Ga$_{1-x}$As SLs allowed us to calculate the spectra of the coefficient of optical gain as functions of the pumping density in the region of nonlinear effects.

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ЛЮМІНЕСЦЕНТНІ ВЛАСТИВОСТІ ПРИПОВЕРХНЕВИХ НАПІВПРОВІДНИКОВИХ ШАРІВ ТА КВАНТОВИХ НАДҐРАТОК

В.Г. Литовченко, Д.В. Корбутяк

Р е з ю м е

Наведено нові результати люмінесцентних досліджень квантових надґраток (НҐ) GaAs/AlAs I-го та II-го роду, а також меж поділу гетероструктур GaAs/AlAs з використанням імпульсних фемтосекундних джерел збудження. Проаналізовано особливості кінетики фотолюмінесценції НҐ GaAs/AlAs з різними ширинами квантових ям (GaAs) та бар'єрів (AlAs). При високих рівнях збудження у квазіпрямозонних НҐ формується електронно-діркова плазма, концентрація носіїв заряду в якій більш ніж на порядок переви-

щує відповідне значення для об'ємних кристалів GaAs. Дослідження спектрів спонтанного вимушеного випромінювання електронно-діркової плазми в НҐ GaAs/AlAs дозволило розрахувати спектри коефіцієнтів оптичного підсилення залежно від густини оптичної накачки в області нелінійних ефектів.