

TUNABLE PHOTOLUMINESCENCE SPECTRA OF DOPED SEMICONDUCTOR SUPERLATTICES***V. K. Kononenko,^{a**} H. W. Kunert,^b I. S. Manak,^c
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Taking into account the density state tails appearing due to fluctuations of impurity concentrations, the spontaneous emission spectra of doped semiconductor superlattices are calculated. In the framework of the model developed, the explanation of the experimentally observed longwave edge and the shift of the photoluminescence spectra with increase in the excitation level and temperature is given. The role of the defects formed on α -irradiation is discussed, and the lifetime of current carriers is evaluated depending on the design parameters and excitation conditions of the GaAs doped superlattices.

Keywords: *doped superlattice, photoluminescence spectrum, lifetime, α -irradiation.*

Introduction. Doped superlattices with the structure of the *n-i-p-i* crystal type belong to semiconductor materials with a tunable energy spectrum. They are grown in the process of periodic doping of a semiconductor crystal with donor and acceptor impurities. When the degree of doping or the level of excitation of such crystals changes, the structure of the energy levels in them undergoes transformation and, accordingly, their absorption and luminescence spectra are transformed [1]. Introduction of defects by irradiation, for example, by α particles, and their recharging in the process of thermal treatment of the structures can also change the potential relief of the superlattice, which must reflect on the luminescence spectra. Radiation defects markedly influence the lifetime of the current carriers and the luminescence quantum yield. Therefore, analysis of the observed changes in the emission spectra of doped superlattices and comparison with predicted spectra allow one to determine the structure parameters of the superlattices and elucidate the role of the appearing inhomogeneities, the nature of the defects, and their influence on the processes of recombination and transfer of current carriers. Moreover, investigation of the change in the optical and electric characteristics of the doped superlattices on introduction of defects is of importance in determining the efficiency of the work of various optoelectronic devices based on them under the conditions of radiative irradiation [2].

In the present work, the spontaneous emission spectra of doped superlattices based on GaAs have been calculated with allowance for the density-state tails arising as a result of fluctuations of the concentration of impurities. The longwave wing and shift observed in the photoluminescence spectra on increase in the excitation level and temperature are explained within the framework of the model developed, and the role of the defects formed under α -irradiation is discussed.

Properties of Doped Superlattices. Because of the spatial separation of electrons and holes in *n-i-p-i*-crystals, in contrast to bulk crystals, the screening of the fluctuating electrostatic potential is brought about separately by electrons in the *n*-regions and by holes in the *p*-regions [3]. When a doped superlattice is excited, the screening lengths decrease, and this changes the distribution of the energy levels and the emission spectrum of a semiconductor.

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The density-state tails play a leading role in compensated strongly doped superlattices, especially at low excitation levels [4]. This is explained by the fact that under these conditions the screening lengths in the n - and p -regions are fairly long and, therefore, in the luminescence spectrum there is a wing which extends far to the longwave region. In noncompensated doped superlattices, the density-state tail exerts the main influence on the shape of the luminescence spectrum only in the n - or p -type regions. The distribution of impurities and the thickness of the n - and p -layers, as well as the dimensions of the intermediate undoped i -regions, determine the shape of the potential relief and its depth [5, 6].

To estimate the depth of the potential relief $2\Delta V$, the approximation of the effective width of the space-charge region [7] or the approximation of the effective concentration of ionized impurities can be used [6]. The former approximation is applicable at low enough temperatures for superlattices with a large long spacing. The latter approximation is suitable for short-period superlattices at room and higher temperatures. Introducing the pumping parameter $r = n/N_d d_n$, where n is the two-dimensional concentration of electrons, N_d is the concentration of donors, and d_n is the thickness of the n -layers of the superlattice, in the space-charge approximation [1, 5] we have

$$2\Delta V = 2\Delta V_0 (1 - r) \left(1 - \frac{r}{1 + 4N_a d_i / (N_a + N_d) d_n} \right). \quad (1)$$

In thermodynamic equilibrium, when at $N = N_a d_p - N_d d_n \geq 0$ the concentration $n \approx 0$, the depth of the relief is largest and is equal to

$$2\Delta V_0 = \frac{\pi e^2}{\epsilon} N_d d_n \left(2d_i + \frac{N_a + N_d}{2N_a} d_n \right), \quad (2)$$

where ϵ is the static dielectric constant of the semiconductor.

In the approximation of the effective concentration of impurities, for a superlattice of the p -type ($N \geq 0$) we analogously find

$$2\Delta V = 2\Delta V_0 (1 - r) = \frac{\pi e^2}{\epsilon} \frac{N_d d_n}{2} (d + 2d_i) (1 - r), \quad (3)$$

where $d = d_n + d_p + 2d_i$ is the superlattice spacing. For an uncompensated superlattice, it is worth-while to use a mixed approximation [1], when the potential in the more strongly doped region is described by the effective space charge and the less doped one is described by the effective charge of the impurities. In this case, solving the Poisson equation for a superlattice of the p -type, we obtain

$$2\Delta V = 2\Delta V_0 (1 - r) \left(1 - \frac{r}{1 + (N_a / N_d) (1 + 4d_i / d_n)} \right). \quad (4)$$

Here, $2\Delta V_0$ is determined from expression (2). It is evident that formula (4) gives a value of $2\Delta V$ that is intermediate between those given by (1) and (3). On rather high-power excitation ($r \geq 1$), the potential relief of a doped superlattice corresponds to a practically sinusoidal distribution of the concentration of unlocalized current carriers and, in particular, at $N = 0$ the depth of the relief changes as $r^{-1/3}$ [1].

In the general case, it is necessary to solve self-consistently the Schrödinger and Poisson equations. The calculations show that even though the space-charge distributions can differ markedly in different approximations, the potential relief is practically the same [1]. The approximation of the effective concentration of ionized impurities or the mixed approximation is closest to the exact profile and depth of the potential relief.

The defects which arise, for example, after irradiation by α -particles lead to the appearance of additional levels in the forbidden band of the semiconductor [8–11]. The charges localized on these levels alter the electrostatic-potential distribution and, consequently, distort the shape of the potential relief and also participate in the radiative and radiationless recombination [12]. As a result, the radiation-induced defects not only change the rate of spontaneous re-

TABLE 1. Parameters of Doped Superlattices Based on GaS

Structure	Concentration of donors and acceptors $N_d = N_a, \text{ cm}^{-3}$	Thickness of the n - and p -type layers $d_n = d_p, \text{ nm}$	Thickness of the i -layer $d_i, \text{ nm}$	Depth of the potential relief $2\Delta V_0, \text{ eV}$	Effective width of the forbidden band $E'_{g0}, \text{ eV}$
2	10^{18}	20	0	0.14	1.28
4	10^{18}	40	0	0.58	0.84
6	10^{18}	60	0	1.30	0.12
$4i$	10^{18}	40	40	1.37	0.05
$6i$	10^{18}	60	60	1.41	0.01

combination and the lifetime of the current carriers in the spatially separated quantum wells of the potential relief, but also lead to the appearance of additional luminescence lines.

In analyzing the transformation of emission spectra it is also necessary to take into account the narrowing of the forbidden band with increase in the temperature and the effects of collective interactions [13]. Excitation of a superlattice increases the effective width of the forbidden band because of the decrease in the depth of the potential relief. Correspondingly, the luminescence spectrum shifts to the shortwave region.

In estimating the lifetime of the current carriers, the influence of the following structure parameters of the superlattice on the rate of spontaneous recombination should be taken into account: the concentration of doping impurities, the superlattice spacing, the distribution of defects, and the excitation conditions (i.e., the intensity and frequency of optical pumping and the temperature of the structure). In the quasiequilibrium state of the electrons and holes, the two-dimensional concentration n and the pumping parameter r are uniquely related to the difference between the Fermi quasilevels ΔF . The rate of the optical generation of current carriers in a unit volume of the superlattice for the light flux density P at the frequency ν_{ex} is $kP/h\nu_{\text{ex}}$, where $k(\nu_{\text{ex}})$ is the absorption coefficient and $h\nu_{\text{ex}}$ is the energy of the exciting quanta. If we introduce the effective lifetime of the current carriers τ , then under stationary conditions, $kP/h\nu_{\text{ex}} = (n - n_0)/\tau d = rN_d d_n/\tau d$, where $n_0 \approx 0$ is the equilibrium concentration of the electrons. When there is an additional channel of radiationless recombination with a time constant τ_{nr} (because of the defects), the rate of recombination of the nonequilibrium current carriers at fairly low excitation levels is controlled practically by this channel, since the lifetime constant of the carriers in the case of spontaneous radiative recombination τ_{sp} is much larger [14]. Moreover, a nonlinear process of absorption saturation can appear on a high-power optical excitation [15].

Discussion of Results. Attention is mostly paid to compensated doped superlattices based on GaAs with i -layers (n - i - p - i -structures) and without them (n - p - n - p -structures) [16]. The thicknesses of the n -, p -, and i -type layers were 20, 40, and 60 nm, and the concentrations of the doping impurities Te and Zn reached 10^{18} cm^{-3} (Table 1). The depth of the potential relief $2\Delta V_0$ was preliminarily estimated from formula (2). For structures with a large lattice spacing, containing i -layers, the initial effective width of the forbidden band E'_{g0} was found to be negative, i.e., there is a degenerate distribution of the electrons and holes, and, to determine the potential relief more exactly, it is necessary to take into account the exchange interaction and other collective effects [7, 13]. The values of the depth of the potential relief and the effective width of the forbidden band for these structures, found by self-consistent calculation, are presented in Table 1.

The structures were grown by the method of gaseous-phase epitaxy at a low pressure on GaAs substrates. The number of superlattice spacings is from one to several tens at a total thickness of the structures of the order of 2.4 μm . The photoluminescence spectra were measured on excitation by the Ar^+ laser radiation with an intensity of up to 50 W/cm^2 in the temperature range from 11 to 300 K. We also investigated the influence of the irradiation by α -particles and the thermal annealing of superlattices on the photoluminescence and Raman scattering spectra [17–20].

The calculations of the spontaneous emission spectra of the structures were carried out with allowance for all the main factors which determine the process of radiative recombination in doped superlattices (dimensional quantization, overlapping of the wave functions of electrons and holes, fluctuations of the impurity concentration, screening of

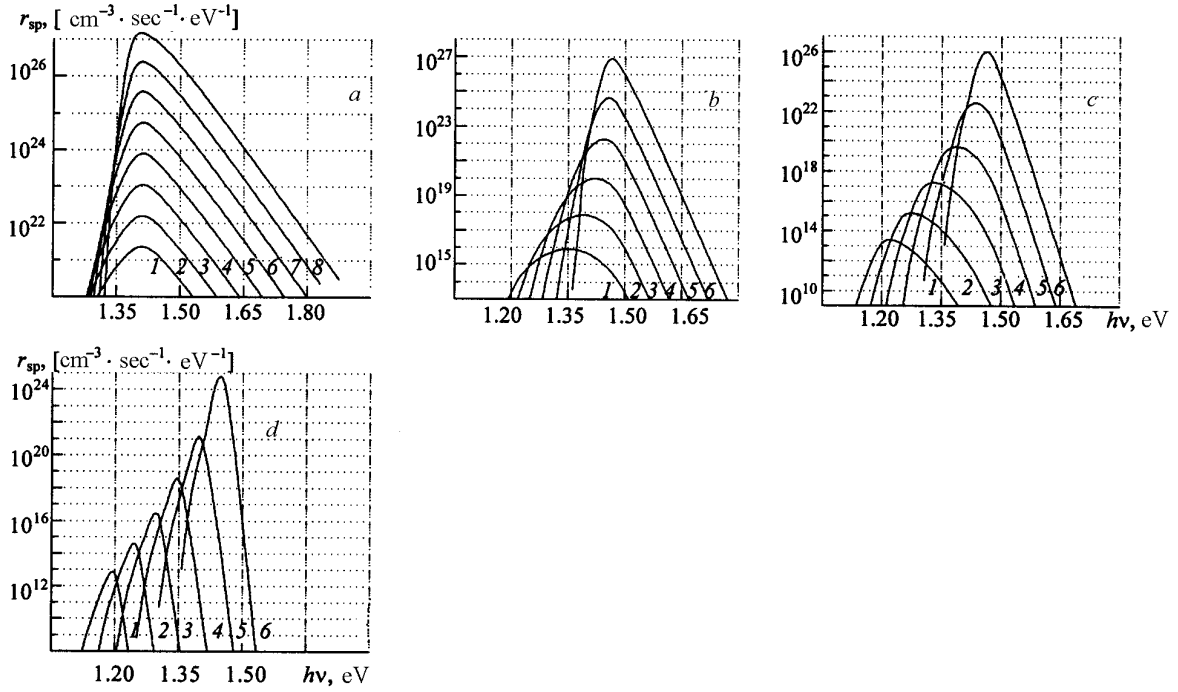


Fig. 1. Spontaneous-emission spectra $r_{sp}(h\nu)$ of a doped superlattice based on GaAs (structure 4i) at different temperatures versus the excitation level (parameters of the superlattice $N_d = N_a = 10^{18} \text{ cm}^{-3}$, $d_n = d_p = d_i = 40 \text{ nm}$): a) $T = 300 \text{ K}$, $\Delta F = 1.00$ (1), 1.05 (2), 1.10 (3), 1.15 (4), 1.20 (5), 1.25 (6), 1.30 (7), and 1.35 eV (8); b) $T = 100 \text{ K}$, $\Delta F = 1.20$ (1), 1.25 (2), 1.30 (3), 1.35 (4), 1.40 (5), and 1.45 eV (6); c) $T = 60 \text{ K}$, $\Delta F = 1.20$ (1), 1.25 (2), 1.30 (3), 1.35 (4), 1.40 (5), and 1.45 eV (6); d) $T = 20 \text{ K}$, $\Delta F = 1.20$ (1), 1.25 (2), 1.30 (3), 1.35 (4), 1.40 (5), and 1.45 eV (6).

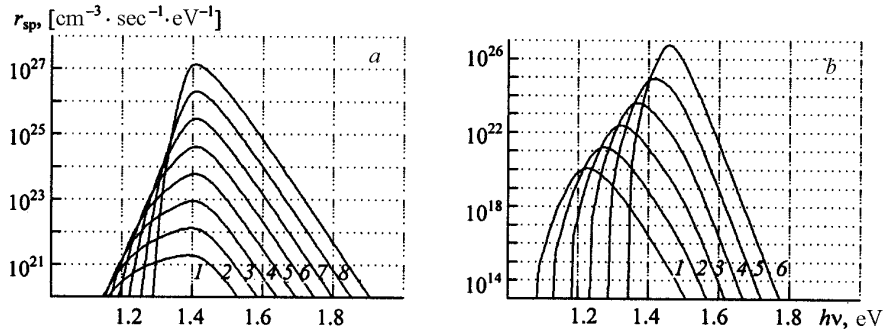


Fig. 2. Spontaneous-emission spectra $r_{sp}(h\nu)$ of a doped superlattice based on GaAs (structure 4) at different temperatures versus the excitation level (parameters of the superlattice $N_d = N_a = 10^{18} \text{ cm}^{-3}$, $d_n = d_p = 40 \text{ nm}$, $d_i = 0$): a) $T = 300 \text{ K}$, $\Delta F = 1.00$ (1), 1.05 (2), 1.10 (3), 1.15 (4), 1.20 (5), 1.25 (6), 1.30 (7), and 1.35 eV (8); b) $T = 100 \text{ K}$, $\Delta F = 1.20$ (1), 1.25 (2), 1.30 (3), 1.35 (4), 1.40 (5), and 1.45 eV (6).

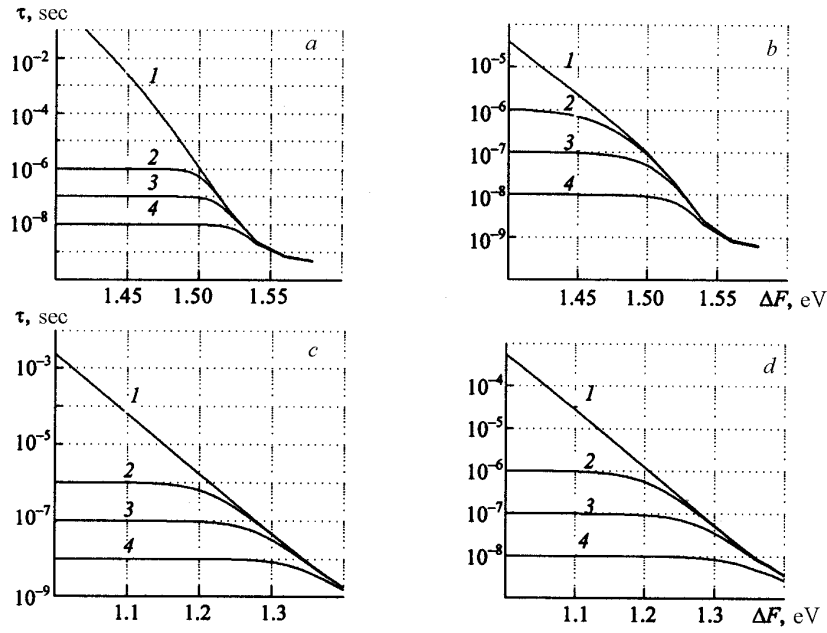


Fig. 3. Dependence of the lifetime of nonequilibrium current carriers τ on the difference between the Fermi quasilevels ΔF in doped superlattices with structures 4*i* (a, c) and 4 (b, d) at the time constant of radiationless recombination $\tau_{nr} = \infty$ (1), 1 μ sec (2), 100 (3), and 10 nsec (4) and the temperature $T = 20$ (a, b) and 300 K (c, d).

the electrostatic potential, and renormalization of the forbidden band). The procedure of calculation of the luminescence spectrum of doped superlattices is described in detail in [1, 4, 13].

The result of calculations for the structures 4*i* and 4 are shown in Figs. 1 and 2. As is seen, at room temperature, the transformation of the spontaneous emission spectrum on increase in the superlattice excitation practically is reduced to the widening of the emission band to the short-wave region, with the maximum of the emission spectrum $h\nu_{\max}$ being located near the forbidden band width of the semiconductor E_g . At nitrogen and lower temperatures, the depth of the potential relief changes more markedly on increase in the pumping, and $h\nu_{\max}$ actually depicts the difference between the Fermi quasilevels ΔF . The effect of the curtailment of the density-state tails because of the change in the screening lengths in the *n*- and *p*-regions is more significant in the structures with *i*-layers.

The estimates of the lifetime of the nonequilibrium current carrier show (Fig. 3) that at room temperature the value of τ_{sp} becomes of the order of 1 μ sec at the pumping level corresponding to $\Delta F = 1.2$ eV. At the nitrogen and lower temperatures this value of τ_{sp} is attained at ΔF close to E_g . Thus, under the conditions of a weak excitation of a superlattice, the rate of recombination of electrons and holes is controlled by the process of the capture of current carriers by the available defects — centers of radiationless recombination, with the role of the defects at the same value of ΔF being more significant in the structures with *i*-layers, because the spatial separation of the electrons and holes is larger in this case.

The change in the photoluminescence spectra versus the temperature of the structures studied is shown in Fig. 4. The structures 4*i* and 6*i* were subjected to irradiation by α -particles with a dose of $3.5 \cdot 10^{13}$ cm^{-2} and were annealed isochronously at 400, 500, and 600°C for an hour. The photoluminescence was excited by the Ar^+ laser radiation of power 50 mW ($h\nu_{\text{ex}} = 2.4$ eV). When the temperature exceeded 80 K, the luminescence intensity S_{ph} decreased markedly. This points to the reduction in the quantum yield of luminescence and decrease in the excitation level and, accordingly, in the concentration of the nonequilibrium current carriers. In both structures, along with the broad tunable radiation band, there is a band in the region of 1.51 eV which is due to the band-to-band transitions, evidently, predominantly in the *i*-layers.

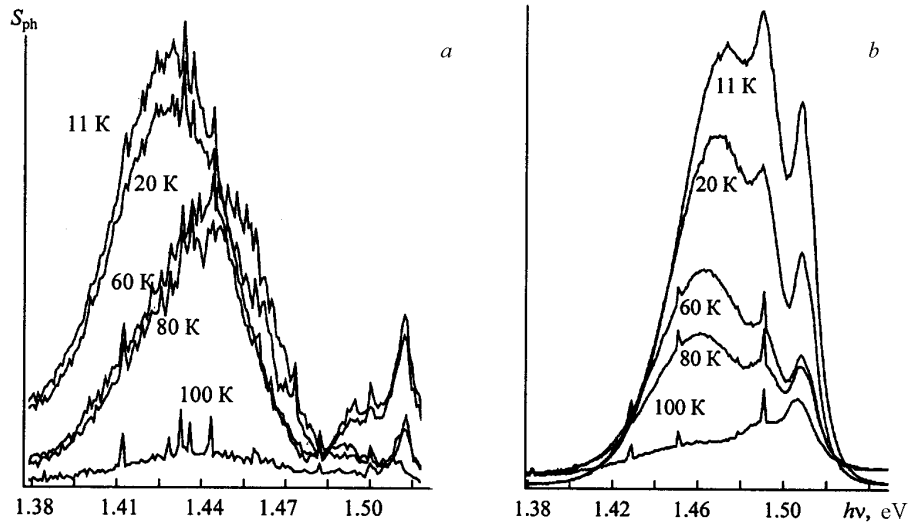


Fig. 4. Temperature dependences of the photoluminescence spectra $S_{ph}(h\nu)$ of doped superlattices based on GaAs with structures 4i (a) and 6i (b) irradiated by α -particles and thermally annealed; $h\nu_{ex} = 2.4$ eV, $P = 50$ W/cm².

Moreover, in the spectra of the structure 6i with superlattice spacing $d = 240$ nm there is a narrow luminescence band with a quantum energy of 1.49 eV, which does not shift when the temperature changes. This new additional band can be due to the radiation defects which appear as a result of irradiation by α -particles. However, these defects are efficiently annealed thermally in the structures with a smaller lattice spacing and practically do not appear in the radiation spectra at $d = 160$ nm, as is seen for the structure 4i. The existence of the additional recombination channel exerts its influence on the level of excitation of nonequilibrium current carriers, which is reflected on the location of the tunable emission band with a change in the temperature and determines the character of the shift of its spectral maximum.

Conclusions. When the degree of annealing and the level of excitation of doped semiconductor superlattices change, the structure of the energy levels undergoes transformation, and, accordingly, their luminescence spectra also transform. Taking into account the density-state tails arising due to the fluctuations of the concentrations of impurities makes it possible to explain the observed longwave wing and shift of the photoluminescence spectrum.

In comparison with the well-known models [21, 22], the model developed for calculating emission spectra more adequately describes all the main features of the changes in the potential relief of doped superlattices on excitation of nonequilibrium current carriers. Because of the spatial separation of the electrons and holes in doped superlattices, the fluctuating electrostatic potential is screened separately by the electrons in the n -regions and by the holes in the p -regions. Upon excitation, the screening lengths decrease, which changes the distribution of the energy levels and the emission spectrum.

Estimates of the lifetime of the current carriers show that the time constant of radiationless recombination depends strongly on the level of excitation of a doped superlattice. Irradiation of compensated doped superlattices based on GaAs by α -particles followed by thermal annealing leads to the appearance of additional lines in the photoluminescence spectra measured in the temperature interval from 11 to 300 K and the stabilization of the lifetime of current carriers at a level of ≈ 10 nsec.

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