NONLINEAR OPTICAL PROCESSES IN DOPED SEMICONDUCTOR SUPERLATTICES[*]

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For doped semiconductor superlattices we have analyzed the effects of absorption saturation and change in the refractive index depending on the level of their excitation and structure parameters. The calculations were carried out with account for the tails of the density of states and screening of the electrostatic potential. It is shown that doped superlattices may display "shading," i.e., the increase in the absorption coefficient at a fixed frequency with increase in light intensity. For δ-doped superlattices a stronger nonmonotonicity of the change in the refractive index with increase in the excitation level in comparison with typical structures can manifest itself.

Keywords: *doped superlattice, absorption saturation, nonlinearity parameter, refractive index.*

Semiconductor structures on doped superlattices have attracted attention because of the widespread possibilities of rearranging their characteristics on both a change in the constructional-technological parameters of the superlattice and structure excitation. Alternate doping of a semiconductor with donor and acceptor impurities leads to modulation of the edges of the conduction and valence bands, resulting in a decrease in the effective energy gap width E'_g, and periodically recurring spatially separated potential wells are formed for electrons and holes. The depth and profile of the potential wells are determined by the thickness of the layers of n- and p-type, d_n and d_p, by the concentration of the donors and acceptors N_d and N_a, by the thickness of undoped i layers d_i, and also by the two-dimensional concentrations of nonequilibrium electrons n and holes p [1].

On excitation of a doped superlattice, the charge of the donor and acceptor impurities is screened by nonequilibrium current carriers, and this causes a decrease in the depth of the potential wells and, correspondingly, an increase in the effective energy gap width. The dependence of E'_g on the excitation level of the structure is responsible for the observed shift to the region of higher energies of the longwave edge of the absorption and luminescence spectra of doped GaAs-based superlattices [2–5].

The change in the properties of doped superlattices with the concentration of nonequilibrium current carriers leads to nonlinear optical effects. In particular, at a given light frequency ν we observe the dependence of the absorption coefficient $k(\nu)$ and refractive index $n_r(\nu)$ on the intensity of light [6, 7]. The absorp-

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tion and refraction spectra can rearrange on both optical excitation and application of electric voltage to the structure. In the present work, we consider the characteristic features of the change in the absorption coefficient and refractive index in a doped superlattice depending on the density of exciting radiation.

Absorption Saturation. As is known, saturation of the absorption power or light amplification is associated with a decrease in the absorption coefficient or amplification of light at a fixed frequency ν with increase in the amplitude of the electromagnetic field strength. In semiconductor systems this effect is caused by the approach of the difference of the Fermi quasilevels ΔF to the energy of photons $h\nu$ on increase in the intensity of monochromatic radiation. The characteristic features of nonlinear absorption or amplification of light were analyzed earlier for bulk semiconductors [8–10] and quantum-dimensional systems [11–13]. For doped semiconductor superlattices this effect has not been studied fully [6, 7, 14]. Moreover, the effect of fluctuations of the electrostatic potential on linear optical processes has not been investigated.

The stationary equation that determines the coupling between the absorption or amplification coefficient k and the density of photons S has the form [8]

$$\frac{\eta' j}{ed} = \frac{R_{\mathrm{lum}}}{\eta_{\mathrm{sp}}} \pm \nu_{\mathrm{g}} k\left(\nu\right) S,\qquad(1)$$

where j is the current density per period of the superlattice d, η' is the injection efficiency, η_{sp} is the quantum yield of luminescence, and ν_{g} is the light velocity in a crystal. In Eq. (1) the second term has the "minus" sign for absorption processes and the "plus" sign for light amplification. The rate of radiative recombination R_{lum} is determined by integration of the velocity of optical transitions $r_{\mathrm{lum}}(h\nu)$ over all the energies of photons. The quantity $r_{\mathrm{lum}}(h\nu)$ is related to the rate of spontaneous recombination $r_{\mathrm{sp}}(h\nu)$ at a given temperature T and level of excitation of the crystal ΔF by the relation [9]

$$r_{\mathrm{lum}}\left(h\nu\right) = \frac{1 - \exp\left(-\dfrac{\Delta F}{kT}\right)}{1 - \exp\left(-\dfrac{h\nu}{kT}\right)} r_{\mathrm{sp}}\left(h\nu\right).$$

The general expression for the rate of spontaneous recombination in the case of direct transitions has the form [1]

$$r_{\mathrm{sp}}\left(h\nu\right) = \frac{A_{c\nu}}{\pi \hbar^2 N_{\mathrm{p}} d} \sum_i m_{ri\perp} \sum_n \sum_m \sum_\nu H_t\left(h\nu - h\nu_{nmi\nu}\right) \left|I_{nmi\nu}\right|^2 f_{\mathrm{e}}\left(E_{cnmi\nu}\right) f_{\mathrm{h}}\left(E_{\nu nmi\nu}\right),\qquad(2)$$

where $A_{c\nu}$ is the Einstein coefficient, N_{p} is the number of periods of the superlattice, $I_{nmi\nu}$ are the overlap integrals of the envelope wave functions of electrons and holes, and $f_{\mathrm{e}}(E_{cnmi\nu})$ and $f_{\mathrm{h}}(E_{\nu nmi\nu})$ are the Fermi–Dirac distribution functions for electrons and holes with the energies

$$E_{cnmi\nu} = E_{c0} + \frac{m_{ri\perp}}{m_c}\left(h\nu - E_{\mathrm{g}}'\right) + \frac{m_{ri\perp}}{m_{\nu i\perp}} E_{cn\nu} - \frac{m_{ri\perp}}{m_c} E_{\nu in\nu},$$

$$E_{\nu nmi\nu} = E_{\nu 0} - \frac{m_{ri\perp}}{m_{\nu i\perp}}\left(h\nu - E_{\mathrm{g}}'\right) + \frac{m_{ri\perp}}{m_{\nu i\perp}} E_{cn\nu} - \frac{m_{ri\perp}}{m_c} E_{\nu im\nu},$$

E_{c0} and $E_{\nu 0}$ are the energies of the conduction band bottom and of the valence band top, m_c, $m_{\nu h\perp}$, and $m_{\nu l\perp}$ are the effective mass of the electrons and the transverse components of the effective masses of heavy

and light holes, respectively, and $m_{ri\perp} = m_c m_{vi\perp}/(m_c + m_{vi\perp})$ is the reduced mass. For GaAs (with the plane orientation {100}) we use $m_c = 0.067m_e$, $m_{vh\perp} = 0.11m_e$, and $m_{vl\perp} = 0.20m_e$. Summation in (2) is carried out over the quantum numbers of the subbands of electrons n, holes m, and minisubbands ν, and over the states of heavy and light holes ($i = h, l$). Transitions between the subbands begin from the energies of the quanta of light $h\nu_{nmi\nu} = E'_g + E_{cn\nu} + E_{vim\nu}$, associated with the effective energy gap width of the doped superlattice $E'_g = E_{c0} - E_{v0}$ and with the energies of the levels of the subbands of electrons $E_{cn\nu}$ and holes $E_{vim\nu}$.

The function $H_i(y)$ is governed by the broadening of the energy spectrum. In the absence of broadening effects in a semiconductor structure, $H_t(y)$ represents the Heaviside step function. For hightly doped superlattices the impurity and the proper zones of a crystal overlap; therefore the tails of the density of states appear [1]. In the case of the Gaussian tail of the density of states, the function $H_t(y)$ has the form $H_t(y) = $ erfc $(-y/\sigma_{cv})/2$ [5], where $\sigma_{cv} = (\sigma_c^2 + \sigma_v^2)^{1/2}$ and σ_c and σ_v are the characteristic parameters of the tails of the density of states in the conduction and valence bands. With increase in the level of excitation of the doped superlattice, the tails of the density of states become shorter due to the screening of the fluctuating impurity potential by nonequilibrium current carriers [1, 15].

The absorption coefficient $k(\nu)$ is related to the rate of spontaneous recombination $r_{sp}(h\nu)$ by the universal relation [9]

$$k(\nu) = \frac{\exp\left(\dfrac{h\nu - \Delta F}{kT}\right) - 1}{\nu_g \rho(h\nu)} r_{sp}(h\nu), \qquad (3)$$

where $\rho(h\nu) = (h\nu)^2 n_r^2/\pi^2 c^2 \hbar^3 \nu_g$ is the density of electromagnetic modes and n_r is the refractive index of the crystal. Using expressions (1)–(3), we find the dependence of the absorption coefficient k on the density of photons S. In the general case it obeys a complex law [8]. If the mean value of the nonlinearity parameter α is taken into account, then the change in the absorption (amplification) coefficient can be described by the simple formula [8, 9]

$$k = \frac{k_0}{1 + h\nu\alpha S}, \qquad (4)$$

in which k_0 is the initial coefficient of absorption (amplification).

To carry out a more detailed analysis of the effects of saturation in specific quantum-dimensional structures, numerical calculations are needed. For doped semiconductor superlattices it is convenient to perform the calculation of nonlinear absorption by the following algorithm [13]. First, by prescribing the difference of the Fermi quasilevels ΔF_0 and assuming the density of photons $S = 0$, it is necessary to determine the corresponding coefficient of absorption k_0 at the exciting light frequency ν and the current density j. If $j = 0$, then $\Delta F_0 = 0$. Increasing ΔF, we find new values of the absorption coefficient k and the recombination rate R_{lum}, and thereafter the corresponding density of photons S is calculated from Eq. (1). Thereupon the procedure is repeated until the difference of the Fermi quasilevels ΔF becomes equal to the energy of photons $h\nu$.

The results of the calculations of absorption saturation for two types of doped GaAs-based superlattices for $j = 0$, $T = 300$ K, and $\eta_{sp} = 0.7$ are presented in Fig. 1. The calculations of $k(\nu)$ and $r_{sp}(h\nu)$ were made in the model of the Gaussian tails of the density of states with account for the screening of the fluctuating impurity potential. As is seen, the absorption coefficient k at a fixed frequency ν exhibits nonmonotonic behavior on increase in the density of photons S. The "shading" effect, i.e., the increase in the absorption coefficient k on increase in the density of photons S, is due to the transformation of the potential energy profile of the doped superlattice, redistribution of the energy levels in quantum wells for electrons and holes,
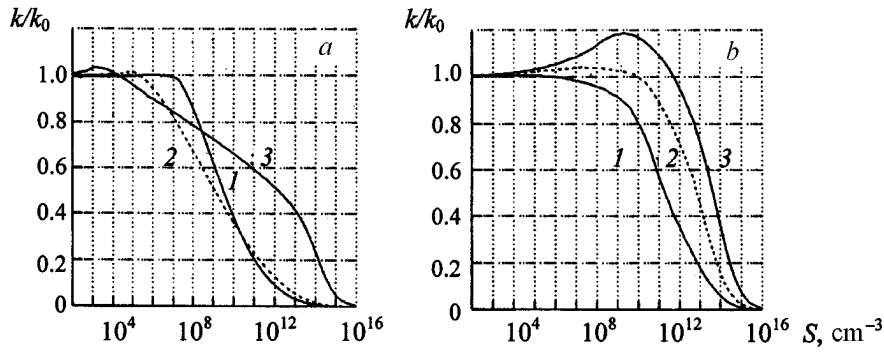
Fig. 1. Absorption saturation for different types of doped superlattices at different energies of photons: a) $N_a = 10^{19}$ cm^{-3}, $N_d = 6\cdot10^{18}$ cm^{-3}, $d_p = 35$ nm, $d_n = 25$ nm, and $d_i = 0$; b) $N_a = N_d = 10^{20}$ cm^{-3}, $d_p = d_n = 1$ nm, $d_i = 8$ nm; 1) $h\nu = 0.8$ eV, $k_0 = 0.1$ (a) and 0.4 cm^{-1} (b); 2) $h\nu = 1.1$ eV, $k_0 = 20$ (a) and 84 cm^{-1} (b); 3) $h\nu = 1.4$ eV, $k_0 = 1537$ (a) and 2437 cm^{-1} (b).

and change in the overlap integrals of wave functions of electrons and holes, and also to the narrowing of the forbidden band.

In the absence of fluctuations of the concentrations of impurities an oscillative change in the absorption coefficient at a fixed frequency of light is observed, depending on the level of excitation of the doped superlattice [6]. In doped superlattices, the fluctuations of the electrostatic potential usually smooth out the stepwise density of states, which leads to smooth absorption and emission spectra [1]. As a result, the function $k(S)$ also becomes smooth, but the region of shading, which most often displays itself at the energy of the photons of the order of the energy gap width of the semiconductor, does not disappear (Fig. 1).

The nonmonotonic character of the absorption saturation is most pronounced for the δ-doped superlattice (Fig. 1b). For bulk semiconductors the shading can be caused by the increase in the absorption by free carriers [8]. However, this process practically does not influence the absorption saturation in doped superlattices, since the coefficient of light absorption by free carriers for the energies of photons of the order of the effective energy gap width of the superlattice does not exceed 5 cm^{-1} [16].

In a wide range of change in the density of photons S, the function $k(S)$ is not described by formula (4) with a constant nonlinearity parameter α. It is more convenient to carry out evaluations of the parameter α using a double logarithmic scale [8] for the dependence of $k_0/k - 1$ on S (Fig. 2). Then the behavior of the curves will show the change in α at the different energies of exciting quanta $h\nu$ depending on the range of the light flux density $h\nu v_g S$.

As is seen, the threshold bleaching fluxes, when the absorption coefficient decreases, e.g., by 1%, for a doped superlattice of p-type is ~10 mW/cm^2 for $h\nu = 0.8$ eV. With increase in $h\nu$ the bleaching thresholds become still smaller. For a δ-doped compensated superlattice the opposite picture is observed: as $h\nu$ increases, the bleaching threshold rises, and at $h\nu = 1.4$ eV it comes to about 2 kW/cm^2. The difference is due to the different character of filling of the states in the conduction and valence bands in optical excitation of superlattices.

As the density of photons S increases, the nonlinearity parameter decreases at any energy of light quanta. This change is attributable to the joint effect of the shortening of the tails of the density of states [10] and to the increase in the effective energy gap width [1]. The latter factor predominates for δ-doped compensated superlattices because of the strong background screening [5].
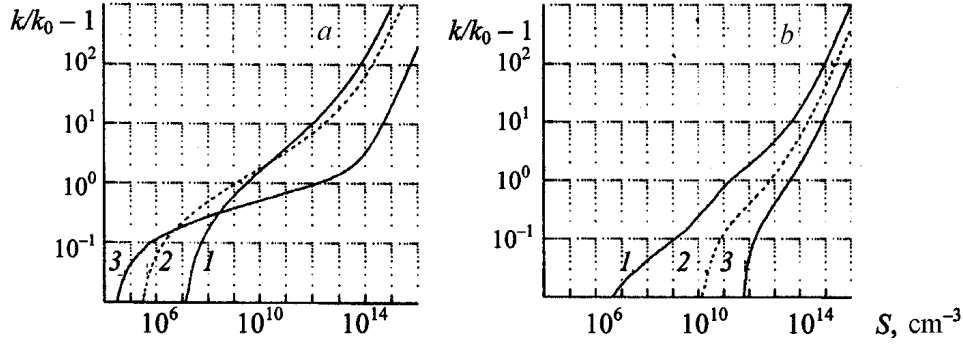
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Fig. 2. Dependence of $k_0/k - 1$ on the density of photons S at different energies of photons for doped superlattices of different types. The parameters of the superlattices are the same as in Fig. 1a and b, curves 1–3, respectively.

For large enough values of S, the change in the absorption coefficient follows formula (4) with a constant value of α [8]. In particular, for the energies of photons $h\nu = 0.8$, 1.1, and 1.4 eV we respectively have $\alpha/v_g = 0.83$, 0.26, and 0.010 cm²/kW for the p-type superlattice (Fig. 2a). For the same energies of photons in the case of a δ-doped superlattice we obtain $\alpha/v_g = 0.10$, 0.028, and 0.0067 cm²/kW (Fig. 2b). As is seen, the values of α decrease with increase in $h\nu$. The light flux density at which the absorption coefficient becomes two times as small varies within wide ranges: from $2W/\text{cm}^2$ in the p-type superlattice to 80 kW/cm² in the δ-doped superlattice.

Nonlinear Dispersion. As is shown in [7], in doped superlattices there is a stronger nonlinear refraction in comparison with bulk semiconductors. The difference in the refractive indices of a doped GaAs-based superlattice and a bulk semiconductor is equal to 0.02. The difference between the refractive indices in excited and unexcited states of doped superlattices based on GaAs attains 0.003 and those based on InAs 0.015. In δ-doped superlattices, still greater nonlinear refraction is possible [17].

The dispersion characteristics of the δ-doped GaAs-based superlattice were calculated with account for screening of the electrostatic potential by current carriers and of the Gaussian tails of the density of states [5]. The refractive index Δn_r at a certain frequency ν_0 was determined from the Kramers–Kronig relation

$$\Delta n_r (\nu_0) = \frac{c}{2\pi^2} \oint \frac{k(\nu)}{\nu^2 - \nu_0^2} \, d\nu \; .$$

Here, the integral is the principal value.

The results of calculations of the absorption coefficient $k(\nu)$ and refractive index $\Delta n_r(\nu)$ are presented in Fig. 3. As is seen, with increase in the excitation level ΔF the maximum of the dispersion curve is shifted to the shortwave region by 100 meV, which corresponds to the shift of the absorption edge because of the increase in the effective energy gap width of the superlattice.

The dependences of the refractive index Δn_r at a fixed frequency of light on the difference of the Fermi quasilevels ΔF are presented in Fig. 4. For the δ-doped superlattice the nonmonotonic character of the change in Δn_r is a stronger function of ΔF, especially for the energies of photons of the order of the energy gap width of the crystal. Here, the change in the refractive index at a fixed frequency with a change in the excitation level of the superlattice for $T = 300$ K can attain 0.02.

Using relation (1), one can easily determine the dependence of Δn_r on the density of exciting photons S. At rather large values of $h\nu$ the nonmonotonic character of the nonlinear refraction $\Delta n_r(S)$ is clearly dis-
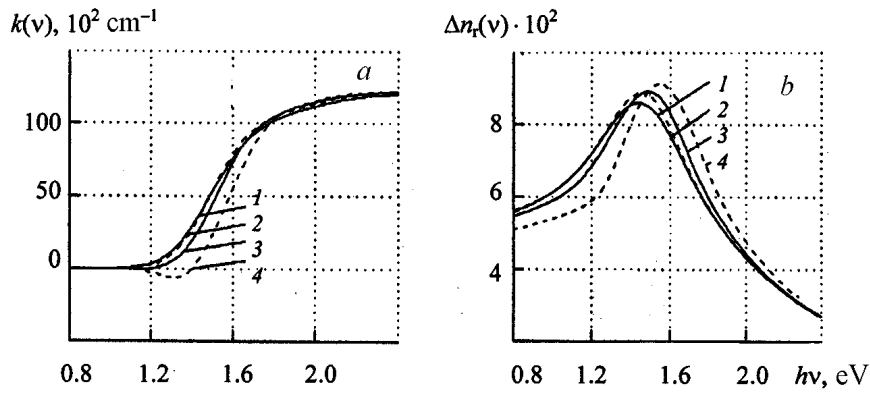
Fig. 3. The spectra of absorption $k(\nu)$ (a) and refraction $\Delta n_r(\nu)$ (b) for the δ-doped superlattice at different excitation levels: $\Delta F = 0.8$ (1), 1.0 (2), 1.2 (3), and 1.4 eV (4). The parameters of the superlattice are the same as in Fig. 1b.
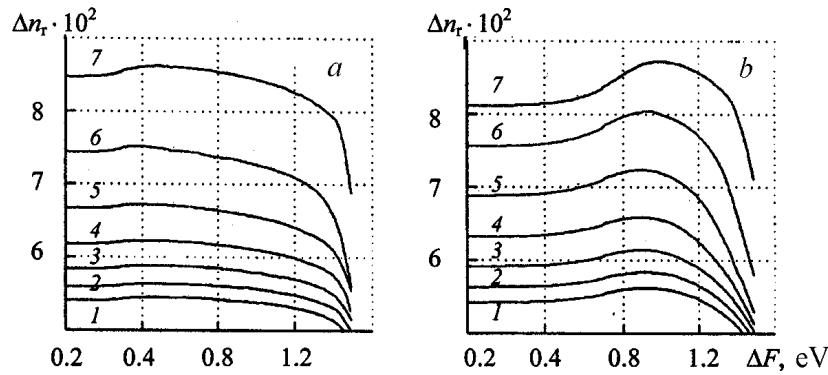

Fig. 4. Change in the refractive index Δn_r at a fixed frequency ν depending on the difference between the Fermi quasilevels ΔF: $h\nu = 0.8$ (1), 0.9 (2), 1.0 (3), 1.1 (4), 1.2 (5), 1.3 (6), and 1.4 eV (7). The parameters of the superlattices are the same as in Fig. 1a and b, respectively.

played. In the limiting case, Δn_r seems to approach a constant value that corresponds to the condition $\Delta F = h\nu$ [8, 13].

The quantity Δn_r should be considered as the addition to the refractive index of the crystal due to the dimensional quantization of levels and the filling of them with nonequilibrium current carriers on excitation of the superlattice. The rather large changes in the absorption coefficient and refractive index in the semiconductor structures with a doped superlattice can be utilized in optical bi- and multistable switches [18]. Moreover, these structures can be built into photon crystals for tuning transmission resonances with a view toward developing low-threshold optical logical elements [19].

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