Current carrier lifetime in doping superlattice crystals

Valerii K. Kononenko^{a*}, Dmitrii V. Ushakov^a, and Herbert W. Kunert^b

^a Stepanov Institute of Physics, National Academy of Sciences of Belarus, Minsk, 220072, Belarus ^b Department of Physics, University of Pretoria, Pretoria, 0002, South Africa

ABSTRACT

It is shown, that the lifetime of nonequilibrium current carriers in the luminescence process in doping superlattice structures changes in a wide range with increasing the excitation level of the crystal. Two effects are important, i. e., (a) low-dimensional character of the carrier distribution and (b) change in the overlap of electron and hole wave functions. At low excitation, non-radiative recombination can play principal cause in the stabilization of the effective lifetime of current carriers. At high excitation, the effective lifetime of current carriers approaches the value in the bulk crystal. The major attention was given to the compensated GaAs doping superlattices with *i*-layers (*n-i-p-i* crystals) and to the structures with no *i*-layers (*n-p-n-p* structures). The layer thickness of *n-*, *p-*, and *i*-type were 20, 40, or 60 nm and the concentrations of the dopants Te and Zn made up to 10^{18} cm⁻³. Photoluminescence spectra and the decay time of the spontaneous emission intensity in the superlattices were measured at the temperature interval from 11 to 300 K. The influence of α -particle irradiation and thermal annealing on the luminescence spectra and the carrier lifetime was also investigated.

Keywords: doping superlattice, current carriers lifetime, luminescence spectrum, non-radiative recombination defect.

I. INTRODUCTION

As an active controllable component in semiconductor structures, doping superlattices are attractive since they are compatible with dense optical and electronic integration and could provide required performance characteristics of the devices. Unique features of doping superlattices, or *n-i-p-i* crystals, are spatial separation of electrons and holes, tunable energy band gap under excitation, increased current carrier lifetime, strong changing the potential profile versus impurity concentrations, and wide variation of electric and optical characteristics due to design parameters or added quantum wells and δ -doped layers.¹

Earlier, from the time-resolved observation of luminescence in GaAs doping superlattices the lifetime of current carriers was determined as a function of the optical excitation power.² The analysis of changes in the total and radiative lifetimes was performed in the model with no the k-selection rule and without the taking into account various possible broadening mechanisms. In spite of this, the predictions agree well with the experimentally observed increasing of the lifetimes over several orders of magnitude during the red shift of the luminescence spectra.

In this paper, processes connected with radiative recombination of nonequilibrium current carriers in the doping superlattice GaAs crystals are analyzed. Data of self-consistent calculations of the electrostatic potential profile in the superlattices are presented and transformation of electron energy levels and wave functions and variation of the overlap of electron and hole wave functions under excitation and in dependence on the superlattice design parameters are examined in details. Influence of high doping on the energy spectrum and electron-hole recombination in n-i-p-i crystals is determined taking into account the Gaussian fluctuations of the impurity concentrations. Generally, the fluctuations of dopant concentrations, correlation in the impurity distribution, and screening of the electrostatic potential have an effect independently in n- and p-regions. Account of the appearing tails of the density of states allows describing adequately the long-wavelength edge in the luminescence spectra and their transformation under excitation and also other unusual peculiarities in optical and electric characteristics of doping superlattices.

^{*} Further author information -

V.K.K. (correspondence): Email: lavik@dragon.bas-net.by; Telephone: 375(017)284-04-35; Fax: 375(017)284-08-79

D.V.U.: Email: ushakovdv@bsu.by

H.W.K.: Email: hkunert@nsnper1.up.ac.za

Solid State Crystals 2002: Crystalline Materials for Optoelectronics, Jaroslaw Rutkowski, Antoni Rogalski, Editors, Proceedings of SPIE Vol. 5136 (2003) © 2003 SPIE · 0277-786X/03/\$15.00

2. RESULTS

The major attention was given to the compensated GaAs doping superlattices with *i*-layers (*n-i-p-i* crystals) and to the structures with no *i*-layers (*n-p-n-p* structures). The layer thickness of *n-*, *p-*, and *i*-type were 20, 40, or 60 nm and the concentrations of the dopants Te and Zn made up to 10^{18} cm⁻³ (Table 1). The values of the potential relief depth $2\Delta V_0$ and effective energy band gap of the superlattice structures E'_g given in Table 1 have been obtained by the self-consistent calculation procedure.

Photoluminescence spectra of the superlattices were measured at varied temperature T in a wide interval from 11 to 300 K. The influence of α -particle irradiation and thermal annealing on the luminescence spectra was also investigated.³

Sample number	Donor and acceptor concentrations $N_d = N_a$, cm ⁻³	Thickness of <i>n</i> - and <i>p</i> -layers $d_n = d_p$, nm	Thickness of <i>i</i> -layers <i>d_i</i> , nm	Potential relief depth $2\Delta V_0$, eV	Effective band gap E'_g , 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
4 <i>i</i>	10 ¹⁸	40	40	1.37	0.05
6 <i>i</i>	10 ¹⁸	60	60	1.41	0.01

Table 1. Parameters of the GaAs doping superlattice structures. T = 300 K.

As the analysis of the rate of optical transitions shows, the lifetime of nonequilibrium current carriers in the luminescence process changes in a wide range with increasing the excitation level of the superlattices. Two effects are important, i. e., (a) low-dimensional character of the carrier distribution and (b) change in the overlap of electron and hole wave functions. At low excitation, non-radiative recombination can play principal cause in the stabilization of the effective lifetime of current carriers. At high excitation, the effective lifetime of current carriers approaches the value in the bulk crystal, i. e., close to 0.7 ns (GaAs). Depending on the impurity concentrations and layer thickness, the effective band gap of doping superlattices can be designed as low as for an inverted arrangement of the intrinsic band edges. Accordingly, the emission spectra and carrier lifetime are significantly varied by design parameters and with increasing the optical excitation.

In the general case, the radiative lifetime of current carriers τ_{sp} in doping superlattice crystals is defined as

$$\pi_{sp} = \frac{n - n_0}{dP_{sp} \left[1 - \exp\left(-\Delta F / kT\right) \right]},\tag{1}$$

where *n* is the two-dimensional concentration of electrons, n_0 is the equilibrium value of the concentration, *d* is the period of the superlattice, R_{sp} is the spontaneous recombination rate, ΔF is the difference of the quasi-Fermi levels, *T* is the temperature of the crystal. Here, it is assumed that the energy of emitted quanta *h*v exceeds the thermal energy *kT* and the width of the spontaneous emission band is comparatively narrow.

The concentration of electrons is given by summation over the whole subbands, i. e.,

$$n = \frac{m_c}{2\pi\hbar^2 N_p} \sum_n \sum_{\nu} \int \operatorname{erfc}\left(\frac{E_{c0} + E_{cn\nu} - E}{\sigma_c}\right) f_e(E) dE \,. \tag{2}$$

Here m_c is the effective mass of electrons, N_p is the number of the superlattice periods, E_{c0} is the bottom of the conduction band in *n*-layers, E_{cnv} is the ground state for the subband with the quantum number *n* and the miniband with the quantum number v, $f_e(E)$ is the distribution (dependent on the quasi-Fermi level F_e) of electrons over the energies *E*. A similar expression with corresponding parameters ($m_{vi}, E_{vinv}, \sigma_v, F_h$) is written for the concentration of heavy and light holes (i = h, l) in the subbands in *p*-layers. Effects of fluctuations of concentrations of impurities are taking into account through the broadening parameters of the density state tails σ_c and σ_v .^{1, 4} Under excitation of the superlattice the screening of the electrostatic potential becomes more effective that influences on the energy tail distribution and on the population in the subbands.⁵ The spectrum of the spontaneous emission recombination in doping superlattices is also modeling taking into account appeared density state tails (owing to fluctuations of the impurity concentrations in the *n*- and *p*-type layers).^{1, 4} The rate of spontaneous recombination can be calculated in the case of direct transitions and in the model with no the k-selection rule as well. In the first approach, the spectrum of the spontaneous emission recombination is given as follows

$$r_{sp}(h\nu) = \frac{A_{c\nu}}{2\pi\hbar^2 N_p d} \sum_{i} m_{ri\perp} \sum_{n} \sum_{m} \sum_{\nu} \operatorname{erfc}\left(-\frac{h\nu - h\nu_{nmi\nu}}{\sigma_{c\nu}}\right) |I_{nmi\nu}|^2 f_e(E_{cnmi\nu}) f_h(E_{\nu nmi\nu}).$$
(3)

Here, A_{cv} is the Einstein coefficient, $m_{ri\perp}$ is the reduced mass relative to transverse components of the hole masses, $hv_{nmiv} = E'_g + E_{cnv} + E_{vimv}$, $\sigma_{cv} = \sqrt{\sigma_c^2 + \sigma_v^2}$, I_{nmiv} is the overlap integral of the envelope wave functions of electrons and holes, $f_e(E_{cnmiv})$ and $f_h(E_{vnmiv})$ are the Fermi-Dirac distribution functions for electrons and holes having the energies determined by the rule of conservation of the wave vector of electrons at the optical transitions. Therewith, $E_{cnmiv} - E_{vnimv} = hv$. Here it is assumed that the energy fluctuations of the edges of the conduction band and valence band in n- and p-regions of the doping superlattice are independent and for the energy levels E_{cnmiv} and E_{vnmiv} in the bands, between which optical transitions occur, average values are taken. Temperature dependence of the energy band gap of the semiconductor E_g is also included according to the empirical Varshni relation.

In the case of optical transitions without selection rules on the electron wave vector, the spectral rate of spontaneous emission recombination is described as follows

$$r_{sp}(h\nu) = \frac{A}{d^2} \sum_{i} \sum_{n} \sum_{m} \int \rho_{cn}(E) \rho_{vim}(E - h\nu) |I_{nmi}|^2 f_e(E) f_h(E - h\nu) dE, \qquad (4)$$

where $A = 4\pi a_0^2 dA_{cv}$ is the probability of optical transitions with no the k-selection rule, a_0 is the effective Bohr radius of the impurity, ρ_{cn} and ρ_{vim} are the densities of states in the subbands of electrons and heavy and light holes. Here, the dependence of the probability A on the characteristic parameter of the superlattice, i. e., on the superlattice period d is taken into account.⁶ Such a dependence can be important for short-period superlattices where $d \le 20$ nm and close with $a_0 \approx 2$ nm. In addition, the averaging over the whole quantum numbers v of the minibands is carried out.

The rate of spontaneous radiative recombination R_{sp} is obtained as an integral sum of $r_{sp}(hv)$ over the energy hv of all emitted quanta. Then, using Eq. (1) the radiative lifetime of current carriers τ_{sp} can be evaluated. If included existing, in general, channels of non-radiative recombination, the effective lifetime of current carriers τ is determined through the relation

$$\tau = \frac{\tau_{sp}\tau_{nr}}{\tau_{sp} + \tau_{nr}},\tag{5}$$

where τ_{nr} is the non-radiative lifetime of the carriers, or via the definition $\tau = \eta_{sp} \tau_{sp}$, where η_{sp} is the quantum efficiency. As a rule, it is assumed that τ_{nr} does not change with the excitation of the crystal. But, in the general case non-radiative process exhibits, as the radiative recombination, bimolecular or more complex character.^{7, 8} After irradiation treatment of the doping superlattices a set of different recombination centers appear.^{3, 9} In conditions of powerful excitation of the superlattices the Auger recombination can be an essential non-radiative process.¹⁰

As seen in Eqs. (3) and (4), the rate of spontaneous emission recombination in doping superlattices in a great degree is determined by the strength of overlapping the electron and hole wave functions. The average meaning of the squared overlap integral is given by

$$|I|^{2} = \frac{1}{np} \sum_{i} \sum_{n} \sum_{m} n_{n} p_{mi} |I_{nmi}|^{2}, \qquad (6)$$

where n_n and p_{mi} are the concentrations of electrons and holes (i = h, l) in the subbands with the quantum numbers n and m respectively. Then, the rate of spontaneous recombination R_{sp} is estimated based on Eq. (4) in the form

$$R_{sp} = \frac{A}{d^2} n(n+N) |I|^2, \qquad (7)$$

where p = n + N, $N = N_a d_p - N_d d_n \ge 0$. If assuming $n_0 \approx 0$, the expression for the radiative lifetime of carriers becomes as follows

$$\tau_{sp} = \frac{1}{4\pi a_0^2 A_{cv} (n+N) |I|^2} \,. \tag{8}$$

For the average value $|I|^2$ of the squared overlap integral it can be roughly assumed the value for the transitions between the ground subbands of electrons and holes.^{8, 11, 12} Functional dependence $|I|^2$ on the excitation level of the superlattice is approximately described by

$$\left|I\right|^{2} = 2\frac{\sqrt{E_{c1}E_{\nu1}}}{E_{c1} + E_{\nu1}}\exp\left(-2\frac{2\Delta V}{E_{c1} + E_{\nu1}}\right),\tag{9}$$

where E_{c1} and $E_{\nu 1}$ are the initial states in the ground subbands of electrons and holes. For simplicity, $E_{\nu 1}$ is taken as the average of the energies $E_{\nu h1}$ and $E_{\nu l1}$ for the subbands of heavy and light holes. The values of E_{c1} , $E_{\nu h1}$, and $E_{\nu l1}$ are determined by the donor and acceptor concentrations and effective masses of carriers and slightly excitation dependent.⁸ The depth of the potential relief $2\Delta V$ is directly related to the excitation factor $r = n/N_d d_n$. For compensated or *p*-type doping superlattices, e. g., $2\Delta V$ is proportional to (1 - r) (within the effective concentrations of ionized impurities approximation).¹ Thus, the optical transitions with no the k-selection rule decay by a complex process.

Obviously that in a quasi-exponential and hyperbolic decay process of the luminescence intensity the time constant τ_d will vary with the time t according to the function

$$\tau_{d} = \frac{\tau_{nr}}{2} \left(1 - \frac{\tau_{nr}}{\tau_{nr} + \tau_{sp0}} e^{-t/\tau_{nr}} \right), \tag{10}$$

where τ_{sp0} is the initial value of the radiative lifetime of current carriers. As seen, the decay time constant τ_d increases during decay of the excited carriers from the initial value $\tau_0 = \tau_{sp0} \tau_{nr}/2(\tau_{sp0} + \tau_{nr})$ to the limit quantity $\tau_{nr}/2$. With decreasing the excitation power the initial value of the decay time τ_0 must approach the limit time constant. In doping superlattices, the decay of the luminescence intensity occurs more complex since during decay of excited electrons and holes the overlap of their wave functions can vary significantly.¹¹

Results of calculations of the effective lifetime of current carriers τ in the case of conservation of the electron wave vector are presented in Fig. 1. Evaluations of the lifetime of nonequilibrium current carriers show that at room temperature the value of τ_{sp} becomes of the order of 1 µs at the excitation level corresponding to $\Delta F = 1.2$ V. At liquid nitrogen and lower temperatures the same value of τ_{sp} reaches at ΔF close to E_g . Thus, in the conditions of weak excitation of the superlattice the rate of recombination of electrons and holes is controlled, in general, by a process of the trapping of current carriers by existing defects, which serve as centers of non-radiative recombination. The effect of stabilization of the effective lifetime of current carriers in homogeneously doped superlattices is more pronounced in comparison with δ -doped short-period superlattices.¹³ Therewith, the role of defects at the same value of ΔF occurs more essential in the structures with *i*-layers in which case the spatial removal of electrons and holes is larger. The measured at low temperatures decay time constant τ_d for tunable photoluminescence band in the α -irradiated GaAs doping superlattices lies in the interval 7 to 12 ns (sample 4).³ It leads to the conclusion that non-radiative recombination is the controlling process of the decay of optically excited carriers in the treated doping superlattice structures and the value of τ_{nr} is of the order of 10 ns.



Fig. 1. Dependence of the nonequilibrium current carriers lifetime τ on the quasi-Fermi level difference ΔF in doping superlattices with structures (a, c, e, i) 4*i* and (b, d, f, j) 4 at different values of the non-radiative recombination time constant (1) $\tau_{nr} = \infty$, (2) $\tau_{nr} = 1 \mu s$, (3) $\tau_{nr} = 100$ ns, and (4) $\tau_{nr} = 10$ ns and temperatures (a, b) T = 20 K, (c, d) T = 80 K, (e, f) T = 150 K, and (i, j) T = 300 K.

 $\Delta F(eV)$

 $\Delta F(eV)$

 $\Delta F(eV)$

 $\Delta F(eV)$

3. CONCLUSIONS

Doping superlattices or *n-i-p-i* crystals belong to the class of tunable semiconductor materials. The changing of the doping and excitation level transforms the structure of the energy levels and, accordingly, the spectra of luminescence. The suggested approach takes into account the density state tails appeared due to fluctuations of impurity concentrations and gives possibility to explain the observed intensity distribution at the long-wavelength edge and photoluminescence spectrum shift. In comparison with known models,^{14, 15} the developed modeling of the emission spectra includes main peculiarities of the potential relief and electronic state spectrum behaviors under excitation. Due to spatial separation of electrons and holes, the screening of the fluctuated electrostatic potential takes place independently in n- and p-regions. Under excitation, the shortening of the screening lengths changes distribution of energy levels and influences on the emission spectra.

The evaluations of the lifetime of current carriers show that the radiative recombination time constant occurs to be strongly dependent on the level of excitation of the doping superlattice. Irradiation with α -particles and following isochronal thermal annealing of the compensated GaAs doping superlattices result in the appearing in the photoluminescence spectra measured in the temperature interval from 11 to 300 K additional bands and stabilization of the carrier lifetime by the level of the order of 10 ns that can be attributed with the controlling non-radiative recombination process.

ACKNOWLEDGEMENTS

The work was partially supported by the Belarussian Republican Foundation for Fundamental Research under Project No. F02R-095/698. Experimental investigations are developed in the University of Pretoria, South Africa. The presenting author (V. K. K.) would like to thank Professor J. B. Malherbe for hospitality at the Department of Physics at the University of Pretoria where the work was completed.

REFERENCES

- 1. V. K. Kononenko, I. S. Manak, and D. V. Ushakov, "Optoelectronic properties and characteristics of doping superlattices," *Proc. SPIE* **3580**, pp. 10-27, 1998.
- 2. W. Rehm, P. Ruden, G. H. Döhler, and K. Ploog, "Study of time-resolved luminescence in GaAs doping superlattices," *Phys. Rev B* 28, pp. 5937-5942, 1983.
- H. W. Kunert, D. J. Brink, A. Donnadieu, K. Zeaiter, C. Llinares, J. Allegre, G. Leveque, and J. Camassel, "Electronic and structural properties of as-grown and α-particle irradiated GaAs doping superlattices," *Phys. stat. sol.* (b) 210, pp. 699-705, 1998.
- 4. D. V. Ushakov, V. K. Kononenko, and I. S. Manak, "Effects of energy-spectrum broadening in alloyed semiconductor superlattices," J. Appl. Spectrosc. 66, pp. 820-825, 1999.
- 5. V. K. Kononenko and D. V. Ushakov, "Carrier transport and screening in *n-i-p-i* crystals," *Phys. stat. sol.* (b) 211 pp. 743-749, 1999.
- 6. A. A. Afonenko, I. S. Manak, V. A. Shevtsov, and V. K. Kononenko, "Radiation recombination rate in quantum-well structures in the model without k-selection," *Semiconductors*, **31**, pp. 901-907, 1997.
- H. J. Queisser and M. B. Panish, "Luminescence of zinc doped solution grown gallium arsenide," J. Phys. Chem. Solids 28, pp. 1177-1184, 1967.
- 8. G. H. Döhler, "*n-i-p-i* doping superlattices metastable semiconductors with tunable properties," J. Vac. Sci. Technol. B 1, pp. 278-284, 1983.
- 9. V. A. Kozlov and V. V. Kozlovski, "Doping of semiconductors using radiation defects produced by irradiation with protons and alpha particles," *Semiconductors* **35**, pp. 735-761, 2001.
- 10. D. V. Ushakov, V. K. Kononenko, I. S. Manak, and V. E. Shibko, "Influence of the non-radiative recombination on the saturation of absorption in doping superlattices," in *Laser and Optical and Electronic Techniques*, Vol. 6, pp. 76-83, Minsk, 2001.
- 11. .K. Ploog and G. H. Döhler, "Compositional and doping superlattices in III-V semiconductors," Adv. Phys. 32, pp. 285-359, 1983.
- 12. G. H. Döhler and P. P. Ruden, "Theory of absorption in doping superlattices," Phys. Rev. B 30, pp. 5932-5944, 1984.
- 13. V. K. Kononenko, D. V. Ushakov, I. S. Nefedov, V. N. Gusyatnikov, and Ya. A. Morozov, "Control of the transmission of photonic structures with *n-i-p-i* layers," *Proc. LFNM*'2001, 3rd Int. Workshop on Laser and Fiber-Optical Networks Modeling, pp. 97-99, Kharkiv, 2002.
- 14. M. Renn, C. Metzner, and G. H. Döhler, "Effect of random impurity distribution on the luminescence of *n-i-p-i* doping superlattices," *Phys. Rev. B* 48, pp. 11220-11227, 1993.
- 15. C. Metzner, K. Schrüfer, U. Wieser, M. Luber, M. Kneissl, and G. H. Döhler, "Disorder effects on luminescence in δdoped *n-i-p-i* superlattices," *Phys. Rev. B* 51, pp. 5106-5115, 1995.