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Excitonic effects in band-edge luminescence of semiconductors at room temperatures

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Abstract. A theoretical analysis is developed for ascertaining the influence of exciton states on edge luminescence in different semiconductors at high temperatures and high levels of excitation. Screening effects and the Mott transition for excitons have been taken into account using simple relations obtained with a variational method. Dependencies of luminescence quantum efficiency on excitation level are discussed. A mechanism of exciton non-radiative annihilation due to Auger recombination via deep impurity levels is analysed as well. It is shown that the probability of this process decreases with an energy of exciton binding.

Keywords: room-temperature exciton luminescence, high excitation levels, quantum efficiency, exciton nonradiative annihilation, Auger mechanism.

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1. Introduction

As was argued for the first time in 1967 by Nolle [1] at certain conditions the exciton contribution into edge luminescence in semiconductors can be the principal one not only at low (liquid helium or nitrogen) temperatures, but at room temperatures, too. In this case the radiative recombination is suppressed not only by non-radiative multi-phonon processes of energy dissipation, but Auger recombination of carriers, too. Moreover, along with conventional band-to-band Auger recombination of carriers a specific exciton Auger-type annihilation via deep impurity levels contributes to non-radiative dissipation of energy [2-5]. As it was shown in [2,3,6], this recombination mechanism can dominate in silicon, especially at sufficiently large doping levels or high laser excitation. It is expected also, that this mechanism should manifest itself in other semiconductors.

In the present work some properties of edge luminescence have been considered for the case of high temperatures when exciton dissociation time is much less than characteristic exciton annihilation time. If, furthermore, the binding energy of exciton ground state becomes less than thermal energy kT , the isolated lines of free excitons cannot be observed in optical spectra in principle. Nevertheless, even under these conditions the excitons can contribute substantially to the luminescence and determine its intensity at least for concentrations of excess

carriers lower than the critical one corresponding to the exciton Mott transition. We have considered this effect in a simple model, which allows to take into account and evaluate the influence of above mentioned Auger-type mechanism of exciton non-radiative annihilation via deep impurity levels on quantum efficiency of luminescence both in direct- and indirect-gap semiconductors.

2. Formulation of the problem

Since quantum efficiency of radiative recombination should be determined in a wide range of excitation intensities and doping levels, it is necessary to take into account the effect of Coulomb potential screening between electron and hole at sufficiently large concentrations of free carriers. It can be done by the analogy with [7] on the base of variational method. A ground state wave function of the Wannier-Mott exciton can be approximated in the framework of this method by the following expression:

$$\psi(r) = A \exp\left(-\frac{r}{a}\right), \quad (1)$$

where r is the radial coordinate, A is the normalization constant and a is the variational parameter. Potential energy of the electron-hole interaction is taken in a standard form of screened Coulomb potential:

$$V(r) = -\frac{q^2}{\varepsilon \cdot r} \exp\left(-\frac{r}{r_D}\right), \quad (2)$$

where q is the electron (hole) charge, ε is the static dielectric constant, $r_D = \left(\varepsilon kT / 4\pi q^2 (n+p)\right)^{1/2}$ is the screening radius in a nondegenerate case. For further consideration it is also convenient to introduce the following standard designations: the binding energy of the Wannier-Mott exciton ground state in the absence of

screening, $E_0 = -\frac{\mu q^4}{2\varepsilon^2 \hbar^2}$; the corresponding exciton

Bohr radius, $a_B = \frac{\hbar^2 \varepsilon}{\mu q^2}$; the reduced effective exciton

mass, $\mu = m_e \times m_h / (m_e + m_h)$. Then from the minimum condition for the exciton ground state energy with account for screening one can obtain the following equation for variational parameter $\delta = a/a_B$:

$$\delta^3 - (2\beta^2 - 3\beta)\delta^2 - (\beta^3 - 3\beta)\delta + \beta^3 = 0, \quad (3)$$

where $\beta = 2r_D/a_B$.

Finally, the expression for the exciton binding energy in the case of screening reduces to the following one:

$$E_x = E_0 \left(\frac{2}{\delta(1+\delta/\beta)^2} - \frac{1}{\delta^2} \right). \quad (4)$$

The condition $E_x=0$ determines the point of exciton Mott transition. It follows from Eqs (3) and (4) that this transition occurs at $\beta \cong 2.133$. It should be noticed that obtained solution is valid for both direct- and indirect-gap semiconductors. From the screening radius value at the transition point $r_D \cong 1,067a_B$ one can easily obtain the critical concentration of carriers, at which exciton Mott transition takes place. For example, in the framework of our consideration the critical concentration for the case of silicon at $T = 300 K$ turns out to be $\approx 8.3 \cdot 10^{17} \text{ cm}^{-3}$, which is somewhat lower than usually used value $1.03 \cdot 10^{18} \text{ cm}^{-3}$.

It should be noticed also that the condition $\beta \cong 2.133$ for the exciton Mott transition is quite general one and thus is valid at much lower temperatures, too. The only difference, in comparison to the case of high temperatures, is that it is necessary to take into account possible carriers degeneracy, which changes the expression for r_D in appropriate way. For instance, in the case of the degeneracy in electron subsystem one has to use the follow-

ing expression: $r_D^{-2} = \frac{2\sqrt{2}}{\sqrt{\pi}} \frac{q^2 m_n^{3/2} (kT)^{1/2}}{\varepsilon \hbar^3} F'_{1/2}\left(\frac{E_{Fn}}{kT}\right)$,

where m_n is the effective density-of-states mass for electrons, $F'_{1/2}(z)$ the first derivative of the Fermi-Dirac inte-

gral of the one half order over argument, E_{Fn} the Fermi energy for electrons measured from the conduction-band edge.

Expression (4) can be approximated with a good degree of accuracy by straightforward relation:

$$E_x \cong E_0 \left(1 - \frac{2}{\beta}\right)^2 = E_0 \left(1 - \frac{a_B}{r_D}\right)^2, \quad (5)$$

which is typically used in literature.

Fig. 1 illustrates the dependencies of variational parameter δ and exciton binding energy E_x on β . For a comparison, binding energy dependencies are built both with exact formula (4) and approximate one (5). As one can see from this figure the approximate expression (5) actually gives results very close to (4) in the entire β range up to the Mott transition.

In line with approach of [8, 9], neglecting the effects of higher orders [10], the kinetic equations for continuous flow of excess electron-hole pairs generated by monochromatic laser excitation as well as for the flux of

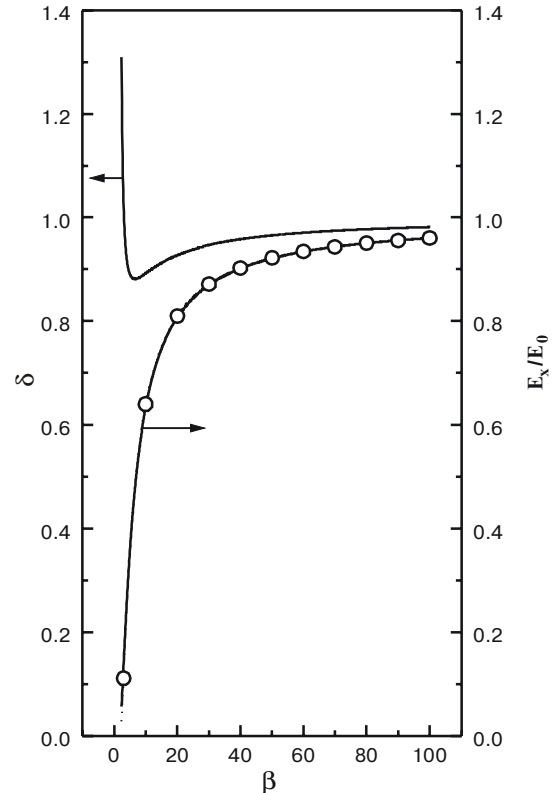


Fig. 1. Dependencies of a variational parameter δ and an exciton binding energy E_x on the screening parameter β . Solid $E_x(\beta)$ curve is built using the exact formula (4), while the dashed with circles one the approximate formula (5). E_0 is the exciton binding energy when screening is absent.

excitons formed due to the Coulomb interaction between carriers can be represented in the following form:

$$\begin{aligned} \frac{\partial \Delta p}{\partial t} = & D_A \frac{\partial^2 \Delta p}{\partial x^2} - \left[\frac{1}{\tau_m} + A_i (n_0 + \Delta n) + C_n (n_0 + \Delta n)^2 + \right. \\ & \left. + C_p (n_0 + \Delta n) \Delta p \right] \Delta p - \gamma_1 ((n_0 + \Delta n) \Delta p - n^* n_x) + \\ & + \alpha I_0 \exp(-\alpha x), \end{aligned} \quad (6)$$

$$\frac{\partial \Delta n_x}{\partial t} = D_x \frac{\partial^2 \Delta n_x}{\partial x^2} - \frac{\Delta n_x}{\tau_x} + \gamma_1 ((n_0 + \Delta n) \Delta p - n^* n_x), \quad (7)$$

where Δn and Δp are the bulk concentrations of excess electrons and holes inside a semiconductor, τ_m is the Shockley-Reed-Hall life time, A_i is the constant of radiative interband recombination, C_n and C_p are the parameters of band-to-band Auger recombination for electrons and holes respectively, γ_1 is the probability of electron-hole pairs association into the excitons, $n^* = n^{**} \exp(-E_x/kT)$ is the characteristic concentration of excitons in the case of thermodynamic equilibrium, $n^{**} = N_c N_v / N_x$, $N_i = g_i (2\pi m_i kT / h^2)^{3/2}$ is the effective density of states ($i=c, v, x$), g_i is the degeneracy factor, m_i is the effective density-of-state mass in the cases of electrons and holes, $m_x = m_c + m_v$, in the case of exciton, τ_x is the exciton radiative annihilation time, n_x and Δn_x are the total and excess exciton concentrations in crystal, I_0 is the intensity of exciting monochromatic light transmitted through the surface into a crystal in the spectral region of fundamental absorption, α is the absorption coefficient, D_A and D_x are the ambipolar diffusion coefficients for carriers and diffusion coefficient for excitons, respectively.

For definiteness, an n -type semiconductor with an equilibrium bulk electron concentration n_0 is considered. It is supposed also that the hole concentration at an external excitation exceeds substantially equilibrium hole concentration in the absence of excitation and thus $\Delta n_x \cong n_x$.

The characteristic reduced time of exciton annihilation, τ_x , can be found from the rule for the sum of reciprocal times τ_x^r and τ_x^n of radiative and nonradiative exciton annihilation:

$$\frac{1}{\tau_x} = \frac{1}{\tau_x^r} + \frac{1}{\tau_x^n}. \quad (8)$$

The system of equations (6)–(8) should be solved together with the following boundary conditions for the flows:

$$j_p(x=0, d) = -D_A \frac{d\Delta p}{dx} \Big|_{x=0, d} = \mp S_{0(d)} \Delta p(x=0, d), \quad (9)$$

$$\begin{aligned} j_x(x=0, d) = & -D_x \frac{d\Delta n_x}{dx} \Big|_{x=0, d} = \\ = & \mp S^x_{0(d)} \Delta n_x(x=0, d), \end{aligned} \quad (10)$$

where $S_{0(d)}$ and $S^x_{0(d)}$ are the effective rates of electron-hole recombination and exciton annihilation at the surfaces $x=0, d$ of a crystal.

As a rule, at high temperatures, the time of exciton dissociation $(\gamma_1 n^*)^{-1}$ is substantially smaller than exciton annihilation time, τ_x . Indeed, in accordance with estimates of [1,8,11] the dissociation time of free excitons is about 10^{-12} s, which is several orders of magnitude smaller than typical τ_x values in all semiconductor materials. At such conditions it is possible to refer the excitons only in statistical sense, i.e their concentration n_x can become substantial only at high excitation levels due to a quadratic growth of n_x with concentration of nonequilibrium carriers. The relaxation of nonequilibrium excitons has to occur in this case within characteristic time which may differ substantially from exciton annihilation time τ_x . For example, at linear response of a system with regard to major carriers, i.e. when $\Delta n \ll n_0$, the effective relaxation time of excitons turns out to be the same as of excess holes. This is expressed by the following formula:

$$\tau^* = \frac{(n_0 + n^*) \tau_x \tau_r^*}{n_0 \tau_r^* + n^* \tau_x}, \quad (11)$$

where τ_r^* is the effective hole lifetime when all electron-hole recombination channels are taken into account. If $n_0 \ll n^*$ and $\tau_r^* n_0 \ll \tau_x n^*$ then $\tau^* \approx \tau_r^*$. In the case of $n_0 \ll n^*$ and $\tau_r^* n_0 \gg \tau_x n^*$ the effective relaxation time should be $\tau^* \approx \tau_x n^* / n_0$.

In the considered model the approximate solution of Eq. (7) has the following form:

$$n_x \cong \frac{1}{n^*} (n_0 + \Delta n) \Delta p. \quad (12)$$

Then, combining Eqs (6) and (7) as well as taking into account Eq. (12), one can obtain for the case of stationary process at high excitation levels, i.e. when $\Delta n \approx \Delta p > n_0$, the following equation:

$$\begin{aligned} D_A \frac{d^2 \Delta p}{dx^2} - \left[\frac{1}{\tau_m} + A_i (n_0 + \Delta p) + \right. \\ \left. + C_n (n_0 + \Delta p)^2 + C_p (n_0 + \Delta p) \Delta p \right] \Delta p + \\ + \frac{D_x}{n^*} \left[(n_0 + 2\Delta p) \frac{d^2 \Delta p}{dx^2} + 2 \left(\frac{d\Delta p}{dx} \right)^2 \right] - \\ - \frac{1}{n^* \tau_x} (n_0 + \Delta p) \Delta p + \alpha I_0 \exp(-\alpha x) = 0. \end{aligned} \quad (13)$$

Solution of Eq. (13) along with boundary conditions (9) allows to obtain Δp as a function of the intensity of exciting light. It should be emphasized that in the considered case the radiative and nonradiative exciton annihilation, which is described by the fourth term in Eq. (13), actually is equivalent to additional quadratic recombination of electron-hole pairs.

Radiation lifetime of excitons can be evaluated from the principle of detailed equilibrium between absorption and radiation in the region of exciton absorption. For example, the estimates in [1,6] demonstrate that for indirect-gap semiconductors it amounts to $10^{-3} \div 10^{-4}$ s, while for direct-gap semiconductors to $\sim 10^{-9} \div 10^{-10}$ with the tendency to decrease as the band-gap grows.

To estimate exciton nonradiative annihilation time we have used a simple model described earlier in [2,6]. This model is based on the fact that the Wannier-Mott exciton forms a region with a large local carrier density $n_L \sim (4\pi a_B^3/3)^{-1}$. This concentration is sufficiently high to initiate a pronounced Auger recombination process, especially when deep impurities are involved. Really, in conventional materials just this Auger recombination is responsible for exciton nonradiative annihilation. As a result the time of exciton nonradiative annihilation can be expressed by the following formula:

$$\tau_x^n = (n_L G N_t)^{-1}, \quad (14)$$

where G is the constant of Auger recombination process with impurity participation, N_t is the concentration of deep impurities. If the condition

$$\tau_x^r n_L G N_t \ll 1 \quad (15)$$

is realized then the exciton nonradiative recombination can be neglected in comparison with exciton radiative annihilation. It is clear from (15) that the role of exciton nonradiative recombination via Auger mechanism becomes less important with decrease of the concentration of deep impurities. The same effect is expected with n_L

decrease, i.e. when exciton binding energy becomes smaller.

On the other hand, taking into account excitonic transitions, the absorption coefficient in direct-gap semiconductors at the absorption edge can be written in accordance with [12] in the following form:

$$K_x(E_g) = 2\pi B \sqrt{E_0}, \quad (16)$$

where B is the same normalizing factor as in frequency dependence of absorption coefficient when excitonic effects are neglected:

$$K(h\nu) = B(h\nu - E_g)^{1/2}, \quad (17)$$

i.e. $B = 2q^2 \mu^{3/2} / (\epsilon^{1/2} c h^2 m_n)$, c is the light velocity and m_n is the effective electron mass.

The probability of radiative annihilation of excitons is proportional to $K_x(E_g)$. From the relation $K_x(E_g) \tau_x^n \sim \epsilon^{1/2} / \mu m_n$ it follows that the ratio of exciton radiative annihilation to exciton nonradiative recombination turns out to be higher for more «shallow» exciton, i.e. for lower μ and m_n values, which is one confirmation more for the validity of our previous conclusions. At the same time the exciton contribution into absorption in accordance with (16) is larger for excitons with higher binding energy.

In the Table 1 one can find the values of reduced exciton mass, exciton Bohr radius, exciton binding energy as well as n_M , n_L , n^* and n_x at $T=300$ K for different semiconductors. The values of n_x have been obtained for the case of $\Delta p = \Delta n = 10^{17} \text{ cm}^{-3}$.

The data in this Table are based on the information presented in [1,6,13,14].

As one can see from this Table an explicit correspondence exists between some important parameters in different semiconductors in accordance with the above discussed qualitative dependencies. In particular, in such

Table 1.

	E_g , eV	μ/m_0	a_B , nm	E_0 , meV	n_M , 10^{17} cm^{-3}	n_L , 10^{18} cm^{-3}	n^* , 10^{17} cm^{-3}	$n_x = \Delta p^2/n^*$, 10^{16} cm^{-3}
Si	1.12	0.15	4.2	14.7	8.3	3.3	8	1.25
InP	1.35	0.06	10.7	6.0	1.32	0.195	1.46	6.83
GaAs	1.43	0.061	10.4	5.8	1.39	0.21	1.51	6.61
GaP	2.21	0.128	4.14	17	7.3	3.36	2.98	3.35
CdTe	1.47	0.084	6.9	9.6	2.88	0.733	2.11	4.74
CdSe	1.74	0.101	5.57	12	4.27	1.38	2.53	3.95
CdS	2.53	0.143	3.3	25	10.2	6.64	2.59	3.87
ZnTe	2.28	0.13	3.96	19	7.73	3.85	2.83	3.54
ZnSe	2.58	0.137	3.14	28	10.3	7.71	2.16	4.63
ZnO	3.2	0.146	2.87	32	12	10.1	2.04	4.91
α-ZnS	3.8	0.179	2.46	35	17	16	2.46	4.06

semiconductors as GaAs and CdTe, where the internal quantum efficiency of radiative recombination is close to 100 %, excitons are the «shallow» ones and the concentration n_L is relatively small. Values of n_M correspond to critical concentrations of charge carriers at which these exciton Mott transitions take place.

Let us consider the case of high excitation levels providing substantial nonlinear effects in a system of nonequilibrium carriers. Then for uniform absorption of exciting light inside the crystal and negligibly small surface recombination rates at surfaces $x = 0, d$ the Δp dependence on the intensity I_0 is determined by the following formula:

$$\left[\frac{1}{\tau_m} + A_i(n_0 + \Delta p) + C_n(n_0 + \Delta p)^2 + C_p(n_0 + \Delta p)\Delta p \right] \times \times \Delta p + \frac{(n_0 + \Delta p)\Delta p}{\tau_x n^{**}} \exp \left[\frac{E_0}{kT} \left(1 - \left(\frac{2\Delta p}{n_M} \right)^{1/2} \right)^2 \right] = \alpha I_0. \quad (18)$$

The total intensity J_{Total} of band-edge luminescence can be found by calculating the following integral:

$$J_{Total} = \int_0^d \left[A_i + \frac{1}{n^* \tau_x} \right] (n_0 + \Delta p(x)) \Delta p(x) dx, \quad (19)$$

while quantum efficiency η for this process is determined by the formula

$$\eta = \frac{\int_0^d \left[A_i + \frac{1}{n^* \tau_x} \right] (n_0 + \Delta p(x)) \Delta p(x) dx}{I_0 (1 - \exp(-\alpha d))}. \quad (20)$$

It should be pointed out that effect of reabsorption is not taken into account in (19) and (20). Therefore, these equations actually are valid only for a plate with thickness d less than the effective reabsorption length of emitted light in a crystal.

Obtained with Eq. (18) theoretical dependencies of excess electron (hole) concentration on intensity of external laser excitation for the cases of Si, GaAs and InP are shown in Fig. 2. The parameters used to obtain these dependencies are presented in Table 2, which is based on the data of [1,6,15]. As one can see from the figure, high

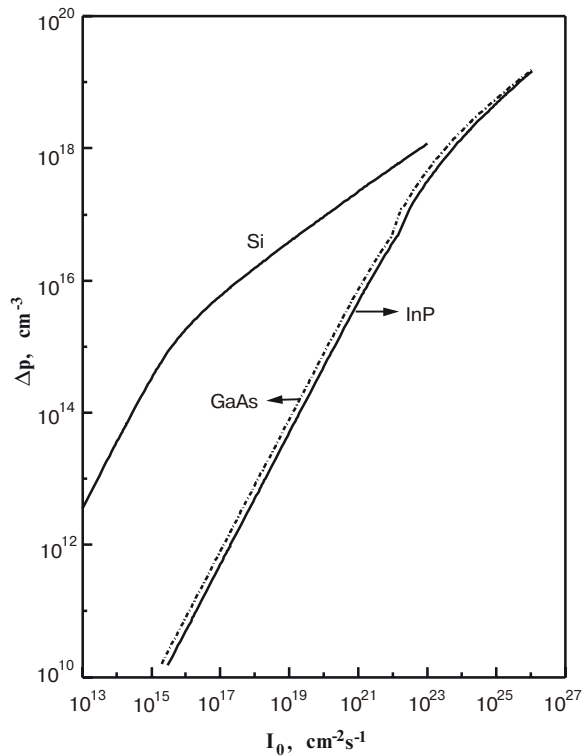


Fig. 2. Concentration of excess electron-hole pairs Δp versus intensity of exciting light I_0 in different semiconductors.

Δp concentrations in Si can be achieved at lower levels of excitation than in GaAs and InP. It is connected with substantially larger value of the Shokley-Reed-Hall lifetime in Si comparing to GaAs and InP.

Figs 3 and 4 demonstrate the dependencies of total photoluminescence intensity and quantum efficiency of radiative recombination on the concentration of excess electron-hole pairs in Si, GaAs and InP. As can be seen from Fig. 3, total photoluminescence intensity in Si is more than three orders of magnitude weaker than in GaAs and InP for the used parameters providing the equivalent values of absorbed radiation in all three materials. In the cases of GaAs and InP an exciton luminescence yields a substantial contribution into the total J value in the region of concentrations $\Delta p < n_M/2$. In the vicinity of the exciton Mott transition, a nonmonotonic behavior in $J(\Delta p)$ dependencies is predicted. It should be particu-

Table 2.

	τ_m, s	τ_x, s	$A_i, cm^3/s$	$C_p, cm^6/s$	$C_n, cm^6/s$	E_0/kT	n^{**}, cm^{-3}	n_M, cm^{-3}	d, mm	α, cm^{-1}
Si	$3.8 \cdot 10^{-2}$	10^{-3}	$1.5 \cdot 10^{-15}$	10^{-31}	$2.8 \cdot 10^{-31} + 2.5 \times 10^{-22} \cdot \Delta p^{-1/2}$	0.567	$1.4 \cdot 10^{18}$	$8.3 \cdot 10^{17}$	100	10
GaAs	$8 \cdot 10^{-9}$	$4 \cdot 10^{-9}$	$2 \cdot 10^{-10}$	$4.6 \cdot 10^{-31}$	$1.6 \cdot 10^{-29}$	0.224	$1.89 \cdot 10^{17}$	$1.39 \cdot 10^{17}$	1	10^3
InP	$5 \cdot 10^{-9}$	$4 \cdot 10^{-9}$	$3.6 \cdot 10^{-10}$	$8.7 \cdot 10^{-30}$	$3.7 \cdot 10^{-31}$	0.232	$1.84 \cdot 10^{17}$	$1.32 \cdot 10^{17}$	1	10^3

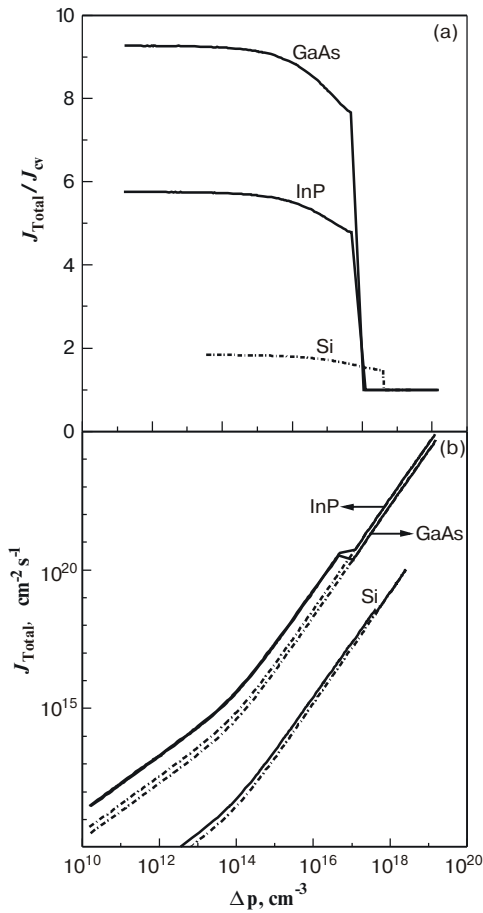


Fig. 3. (a) Ratio of total luminescence intensity J_{Total} including exciton contribution to pure band-to-band luminescence J_{cv} versus concentration Δp of excess electron-hole pairs in Si, GaAs and InP

(b) Dependencies of integrated intensity J_{Total} of photoluminescence on Δp for the same semiconductors. Dashed curves correspond to the case when exciton contribution into luminescence is not taken into account.

larly striking in the cases of GaAs, InP and other direct-gap semiconductors.

Comparing Figs 3 and 4 one can easily understand why silicon is worse light emitter than direct-gap semiconductors: it is due to earlier incorporation of band-to-band Auger recombination with increase of excitation level. Moreover, the larger is the Shokley-Reed-Hall recombination time the earlier this effect takes place. With the used parameters the maximum quantum efficiency for the luminescence in Si is achieved at $\Delta p \approx 10^{15} \text{ cm}^{-3}$. The corresponding intensity of total luminescence is still small in this Δp -region.

Conversely, maximum in quantum efficiency dependencies on Δp for GaAs and InP lies in the Δp -range $10^{18} \div 10^{19} \text{ cm}^{-3}$ where total luminescence intensities also achieve sufficiently high values. Like in Si, this maximum is formed due to a substantial nonlinear increase of

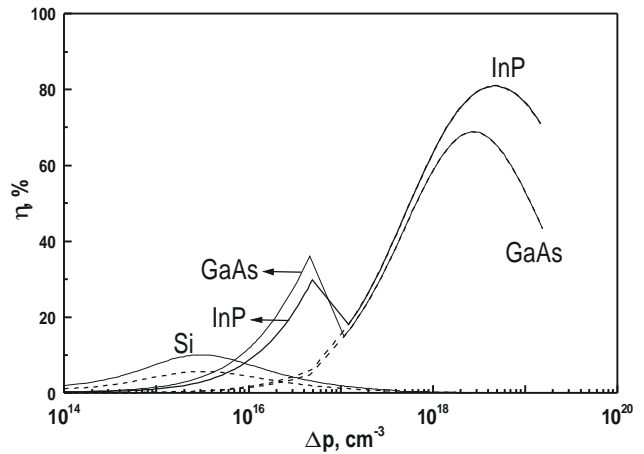


Fig. 4. Quantum efficiency of photoluminescence η versus concentration of excess electron-hole pairs Δp in Si, GaAs and InP. Dashed curves illustrate the case when exciton effects are neglected.

interband Auger recombination. Quantum efficiency in these cases can be as high as 70–80 % in maximum.

To increase the intensity and quantum efficiency of luminescence it would be desirable to obtain a material where the advantages of silicon and direct-gap semiconductors will be combined. Taking into account very high levels of purification and, as a result, extremely large values of the Shokley-Reed-Hall times achieved at the present time in silicon it seems real to use this material as a base. The only task is to obtain in one way or another a direct-gap material on this base which enables to increase the probability of radiative transitions both of band-to-band and exciton types. For example, it can be done by application of uniaxial pressure, or by using nanocrystalline modifications as well as SiGe systems.

One more important point is that maximal quantum efficiency for band-edge luminescence with account for nonradiative channel of exciton annihilation is determined by the following ratio:

$$\eta_{\max} \cong \frac{A_i + 1/n^* \tau_x^r}{A_i + 1/n^* \tau_x^r + n_L G N_t / n^*} \quad (21)$$

Therefore, e.g. for GaAs $\eta_{\max} \approx 1$ in the case of $G N_t \ll 10^{-9} \text{ cm}^3 \cdot \text{s}^{-1}$. I.e. to achieve $\eta \approx 1$ at $N_t \approx 10^{15} \text{ cm}^{-3}$ the Auger constant has to be $G \ll 10^{-24} \text{ cm}^6 \cdot \text{s}^{-1}$, which is quite possible to obtain in this material.

Conclusions

In the present work an attempt has been made to evaluate within simple approximations the role of excitons in band-edge luminescence of semiconductors at high temperatures and high levels of excitation. In fact, only qualitative tendencies are analyzed and thus further theoretical

and experimental investigations are necessary for more detailed quantitative analysis. Nevertheless, it is shown quite definitely that the contribution of exciton radiative transitions into the total luminescence is, as a rule, comparable or even exceeds the contribution of conventional band-to-band radiative transitions even at room temperatures.

Peculiarities in dependencies of quantum efficiency and luminescence intensity on excitation level are considered and analysed both for direct and indirect-gap semiconductors. A contribution of nonradiative channel of exciton annihilation into the total recombination rate is evaluated, too. Obviously, in a number of cases this channel is responsible for the formation in a subsurface region of crystals of the so-called «dead» layer for exciton radiative transitions and exciton luminescence from this region.

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