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HARTREE-FOCK SEMICONDUCTOR BLOCH EQUATIONS AND CHARGE DENSITY CORRELATIONS

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ABSTRACT

Generalized Semiconducor Bloch Equations are derived with using the fluctuation-dissipation theorem. Four operator expectation values like density-density correlators are calculated with account of coherent memory effects. Interactions with mixed plasmon-phonons modes and excitonic effects are taken into account. Comparison with different other theoretical approaches is provided. Numerical calculations of the spontaneous radiation produced by interband multiplasmon recombination of electron-hole pairs are fulfilled in dependence on the temperature and plasma concentration. It is shown that the intensity maximum of spontaneous radiation shifted to the region of the first LO-phonon satellite with increasing of concentration in accordance with experiments [19,21].

Keywords: LO-phonons, electron-hole plasma, correlation effects

1. INTRODUCTION

Correlation effects influencing the optical properties of semiconductors that cannot be described at the basis of Hartree-Fock (HF) approximation are intensively studied at the present time [1-16]. Equations of motion methods [1-6], the Green's function formalism [7-9] or the cumulant expansion method with fluctuation-dissipation theorem [10-16] are successfully used to explain distinctive features of optical spectra of semiconductors.

The recent achievements on correlation effects in semiconductor optics that result as a consequence of the Coulomb interaction and quantum mechanical nature of the light field are discussed by the authors of the work [1]. Scattering processes within the free-particle picture for the propagators of one-particle Green's function include an absorption and emission of plasmon [7-9]. Dynamics-controlled truncation and second Born approximation include correlation contributions representing two electron-hole-pair amplitudes, excitonic populations, and coupled inter- and intraband coherences [1-6]. Correlation and relaxation to the quasi-equilibrium are dominated by a strong coupling of the e-h pairs with plasmons, the longitudinal modes of e-h plasma [10-16].

Multi-plasmon (MP) optical transitions in direct II-VI compound semiconductors ZnSe, CdS and ZnTe at a radiative recombination were investigated for the case of arbitrary density electron-hole plasma (EHP) in a wide excitation power range at various temperatures (4.2 - 355K) [10-16]. A spectroscopic study of MP recombinations was fulfilled using photo and cathodoluminescence (CL) techniques [10-15]. It was found that a new wide radiation band with a thin structure which includes about ten MP replicas appears at high excitation levels in the region of electron-hole plasma light emission. The MP thin structure was shown to be specified by recombination of electrons and holes strongly coupled to the plasmons [10] and it was observed in lattice temperature intervals 40-70K (ZnSe) and 30-120K (ZnTe) [11-13] . It was shown that not only high temperature series of CdS green photoluminescence which is attributed to the free-to-bound optical transitions is strongly coupled with plasmons, but the MP bound-to-bound Donor-Acceptor pairs transitions are actual as well [14]. The red CL of ZnTe doped with oxygen and bound excitons CL bands of ZnSe are composed of phononless line with some plasmon satellites and a great number of LO-phonon replicas [15,16].

In spite of the fact that all these different theories [1-16] deal with just the same problem, the physical mechanisms and results obtained by using these theoretical methods are various in detail drastically. Especially it concerns the

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coherently driven plasmons in high excited semiconductors. Our aim is to derive the equations of motion which allow to include multi-quantum optical processes and excitonic effect into the consideration and which contain the results of another theories. We get the semiconductor Bloch equations (SBE) using the fluctuation-dissipation theorem to calculate four operator expectation values with account of coherent memory effects. This method leads for the interband polarization $P_k(t)$ to energy-conservation $\delta-$functions with arguments of the form $\omega - \omega_g - \omega_k - n\omega_p$, where ω is the photon frequency, ω_p is the frequency of long-wave-length plasmons, involving in process of optical interband transitions and n is a number of plasmons. Here we investigate temperature and plasma concentration dependences of the spontaneous and stimulated radiation with account of multi-quantum transitions.

2. BASIC EQUATIONS

The starting point for our calculations is the Heisenberg equation for the operators. Using the standard many-body Hamiltonian [1-17] for a two-band semiconductor and expanding the macroscopic polarization into a Bloch basis one obtains

$$\begin{aligned} i\hbar\frac{\partial}{\partial t}\hat{P}_{\mathbf{k}_1,\mathbf{k}_2}(t) &= [(e_{\mathbf{k}_2}+h_{\mathbf{k}_1}) - E(t)d_{eh}[\delta_{\mathbf{k}_1,\mathbf{k}_2} - \hat{f}^e_{\mathbf{k}_1,\mathbf{k}_2}(t) - \hat{f}^h_{\mathbf{k}_2,\mathbf{k}_1}(t)] - \\ &\sum_{\mathbf{q}} v_{\mathbf{q}}\{\hat{P}_{\mathbf{k}_1-\mathbf{q},\mathbf{k}_2-\mathbf{q}}(t) + \hat{\rho}_{\mathbf{q}}[\hat{P}_{\mathbf{k}_1,\mathbf{k}_2-\mathbf{q}}(t) - \hat{P}_{\mathbf{k}_1+\mathbf{q},\mathbf{k}_2}(t)]\}. \end{aligned} \quad (1)$$

Operators incoming in Eq. (1) in terms of electron (a^+a) and hole (b^+b) creation and annihilation operators take the next form

$$\hat{P}_{\mathbf{k}_1,\mathbf{k}_2} = b_{-\mathbf{k}_1}a_{\mathbf{k}_2}, \hat{f}^e_{\mathbf{k}_1,\mathbf{k}_2} = a^+_{\mathbf{k}_1}a_{\mathbf{k}_2}, \hat{f}^h_{\mathbf{k}_1,\mathbf{k}_2} = b^+_{-\mathbf{k}_1}b_{-\mathbf{k}_2}$$

Here, standard notations are used:

$$\hat{\rho}_{\mathbf{q}} = \sum_{\mathbf{k}}(b^+_{\mathbf{k}-\ \mathbf{q}}b_{\mathbf{k}} - a^+_{\mathbf{k}-\ \mathbf{q}}a_{\mathbf{k}}) + \hat{\rho}^{ph}_{\mathbf{q}}, v_\kappa = \frac{4\pi e^2}{\varepsilon_\infty V\kappa^2},$$

$e_{\mathbf{k}}$ and $h_{\mathbf{k}}$ are the single-particle energies of an electron in the conduction band and a hole in the valence band. Charge density $\hat{\rho}^{ph}_{\mathbf{q}}$ describes e-h pairs interaction with phonons, $E(t) = E_0\cos\omega t$ is the intensity of classical eletric field. If we let $\mathbf{k}_1 = \mathbf{k}_2 = \mathbf{k}$ and carry out decoupling at the level of a dynamical HF like approximation ($P_{\mathbf{kk}} = P_{\mathbf{k}} =< b_{-\mathbf{k}}a_{\mathbf{k}} >$) we obtain

$$(i\frac{\partial}{\partial t} - \omega_k(t))P_{\mathbf{k}}(t) + \Omega_{\mathbf{k}}(t)(1 - f^e_{\mathbf{k}}(t) - f^h_{\mathbf{k}}(t)) = I_{\mathbf{k}}(t). \quad (2)$$

In this equation the collision term contains direct and exchange, diagonal and off-diagonal contributions

$$\begin{aligned} I_{\mathbf{k}}(t) &= \frac{1}{\hbar}\sum_\kappa v_\kappa\{< \hat{\rho}_{-\kappa}(t)P_{\mathbf{k}-\kappa,\mathbf{k}}(t) > - < \hat{\rho}_\kappa(t)P_{\mathbf{k},\mathbf{k}-\kappa}(t) > + \\ &(f^e_{\mathbf{k}-\kappa}(t) + f^h_{\mathbf{k}-\kappa}(t))P_{\mathbf{k}}(t) - (f^e_{\mathbf{k}}(t) + f^h_{\mathbf{k}}(t))P_{\mathbf{k}-\kappa}(t)\}. \end{aligned} \quad (3)$$

Here, brackets <...>denote averaging with the equilibrium density operator. The effect of the polarization dephasing, screening of interaction potential memory correlation effects as well, are now presented by the function $I_{\mathbf{k}}(t)$. If we let $I_{\mathbf{k}}(t) = 0$, we obtain well known HF semiconductor Bloch equations for polarization [1-16]. There are no contradictions in this point in works [1-16]. In Eq. (2), $\hbar\omega_k(t) = e_{\mathbf{k}}(t) + h_{\mathbf{k}}(t)$ is the exchange renormalized transition energy and $\Omega_{\mathbf{k}}(t)$ is the renormalized field. The effective Rabi energy $\hbar\Omega_{\mathbf{k}}(t)$ contains the dipole energy and HF renormalization. To calculate the function $I_{\mathbf{k}}(t)$ one has to consider the operators $\hat{P}_{\mathbf{k}-\kappa,\mathbf{k}}(t)$ and $\hat{P}_{\mathbf{k},\mathbf{k}-\kappa}(t)$.

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The sum over $\mathbf{q}$ in Eq.(1) includes term with $\mathbf{q} = \kappa$, which has only to be taken into account in Eq.(4) according to the random phase approximation:

$$\hat{P}_{\mathbf{k}-\kappa,\mathbf{k}}(t) = \frac{i}{\hbar} v_\kappa \int_0^t G_{\mathbf{k}-\kappa,\mathbf{k}}(t-t_1)\hat{\rho}_\kappa(t_1)[\hat{P}_{\mathbf{k}-\kappa}(t_1) - \hat{P}_{\mathbf{k}}(t_1)]. \tag{4}$$

This term gives the contribution into Eq.(3) which is proportional to v_κ^2 and diverges as κ^{-4} at $\kappa \to \mathbf{0}$ if we restrict the consideration with second order correction of the perturbation theory. Here, we use the quasi-particle picture

$$G_{\mathbf{k}_1,\mathbf{k}}(t-t_1) = \exp(-\frac{i}{\hbar}\int_{t_1}^t [H_{\mathbf{k}_1}(s) + E_{\mathbf{k}}(s)]ds). \tag{5}$$

Self energies of the quasi-particles $H_{\mathbf{k}}(s), E_{\mathbf{k}}(s)$ have to be calculated selfconsistently.

If we carry out decoupling and use formulas derived we obtain

$$< \hat{\rho}_{-\kappa}(t)P_{\mathbf{k}-\kappa,\mathbf{k}}(t) >= \frac{i}{\hbar} v_\kappa \int_0^t G_{\mathbf{k}-\kappa,\mathbf{k}}(t-t_1) < \hat{\rho}_{-\kappa}(t)\hat{\rho}_\kappa(t_1) > [P_{\mathbf{k}-\kappa}(t_1) - P_{\mathbf{k}}(t_1)]. \tag{6}$$

Correlation function $< \hat{\rho}_{-\kappa}(t)\hat{\rho}_\kappa(t_1) >$can be calculated exactly by using of the fluctuation-dissipation theorem

$$< \hat{\rho}_{-\kappa}(t)\hat{\rho}_\kappa(t_1) >= \frac{\hbar}{\pi v_\kappa}\int_{-\infty}^{\infty} e^{-i\omega(t-t_1)}[n(\omega)+1]\,\mathrm{Im}\{\frac{\varepsilon_\infty}{\varepsilon^*(\kappa,\omega)}\}d\omega. \tag{7}$$

Our consideration is based on the successful concept that the dielectric function $\varepsilon(\kappa,\omega)$ can be computed in the scheme of linear response theory, but the material parameters are depend parametrically on the EHP density, which is assumed to be in a quasi-equilibrium state. We assume that the electromagnetic field strength is arbitrary one. In a similar manner one gets

$$< \hat{\rho}_{\kappa}(t)P_{\mathbf{k},\mathbf{k}-\kappa}(t) >= -\frac{i}{\hbar} v_\kappa \int_0^t G_{\mathbf{k},\mathbf{k}-\kappa}(t-t_1) < \hat{\rho}_{\kappa}(t)\hat{\rho}_{-\kappa}(t_1) > [P_{\mathbf{k}-\kappa}(t_1) - P_{\mathbf{k}}(t_1)]. \tag{8}$$

In Eq.(1) for $\hat{P}_{\mathbf{k},\mathbf{k}-\kappa}(t)$, the only term with $\mathbf{q} = -\kappa$ is taken into account. If we keep all terms proportional to v_κ^2 in the collision integral (3) we obtain

$$I_{\mathbf{k}}(t) = \frac{i}{\pi}\int_0^t dt_1 \sum_\kappa v_\kappa \int_{-\infty}^{\infty} \mathrm{Im}\{\frac{\varepsilon_\infty}{\varepsilon^*(\kappa,\omega)}\}e^{-i\omega(t-t_1)}[C_{\mathbf{k}}(t,t_1) - C_{\mathbf{k}+\kappa}(t,t_1)]d\omega, \tag{9}$$

where

$$C_{\mathbf{k}}(t,t_1) = [\varphi^e_{\mathbf{k}}(t)G_{\mathbf{k},\mathbf{k}-\kappa}(t-t_1) + \varphi^h_{\mathbf{k}}(t)G_{\mathbf{k}-\kappa,\mathbf{k}}(t-t_1)]P_{\mathbf{k}-\kappa}(t_1),$$

and

$$\varphi^e_{\mathbf{k}}(t) = [n(\omega) + 1 - f^e_{\mathbf{k}}(t)], n(\omega) = [\exp(\frac{h\omega}{k_0 T_e}) - 1]^{-1}.$$

We can use Eq. (2) and Eq. (9) to determine the polarization $P_{\mathbf{k}}$ selfconsistently . In the quasi-particle picture

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$$P_{\mathbf{k},\mathbf{k}}(t) = \frac{i}{\hbar}\int_0^t G_{\mathbf{k},\mathbf{k}}(t-t_1)E(t_1)d^{eh}(1-f^e_{\mathbf{k}}(t_1)-f^h_{\mathbf{k}}(t_1))dt_1. \quad (10)$$

with function $G_{\mathbf{k},\mathbf{k}}(t-t_1)$ determined by Eq.(5)

Using this scheme, the equations for carrier occupations probabilities may be derived

$$\begin{aligned}\frac{d}{dt}f^a_{\mathbf{k}}(t) &= -\frac{2}{\hbar}\,\mathrm{Im}[\Omega_k(t)P^*_{\mathbf{k}}(t)] + \frac{1}{\pi\hbar}\int_0^t dt_1 \sum_\kappa v_\kappa \int_{-\infty}^{\infty}\mathrm{Im}\{\frac{\varepsilon_\infty}{\varepsilon^*(\kappa,\omega)}\}\\ &\{\mathrm{Re}[P^*_{\mathbf{k}}(t)P_{\mathbf{k}-\kappa}(t_1)\exp[-i\omega(t-t_1)]G_{\mathbf{k}-\kappa,\mathbf{k}}(t-t_1) -\\ &P_{\mathbf{k}}(t_1)P^*_{\mathbf{k}-\kappa}(t)\exp[-i\omega(t-t_1)]G_{\mathbf{k},\mathbf{k}-\kappa}(t-t_1)] +\\ &\cos\omega(t-t_1)[f^e_{\mathbf{k}-\kappa}(t_1)-f^e_{\mathbf{k}}(t_1)][2n(\omega)+1]\,\mathrm{Re}[G_{\mathbf{k},\mathbf{k}-\kappa}(t-t_1)G^*_{\mathbf{k},\mathbf{k}}(t-t_1)]\}. \quad (11)\end{aligned}$$

The first term in the right hand side of Eq(11) describes radiative transitions that either create a e-h pair or annihilate it. At the level of the Born-Markov approximation, where one ignores memory effects and restricts the analysis to two-particle collisions involving electrons, holes, phonons and plasmons, it can be easily seen that second term leads to the well-known Boltzman collision term. And the last term describes the dynamical screening of radiative transitions probability.

The fluctuation-dissipation theorem leads to exact expression (9) no matter what we assume about the inverse dielectric function $\varepsilon^{-1}(\kappa,\omega)$. For numerical calculations we use $\varepsilon^{-1}(\kappa,\omega)$ in the random phase approximation. In this case, using of the fluctuation-dissipation theorem is the simplest way to take into consideration infinitive series in degrees of v_κ of the perturbation theory.

3. RESULTS AND DISCUSSIONS.

In general, according to the Eq.s(9-11) the correlation at time t depends on the evolution of the system from 0 to t. This is the well-known Coulombic memory effect.

At a high level of EHP excitation needed to obtain the plasma gain, one can assume that the memory time is not very long and neglect memory effects in the Markov approximation. To get Markov approximation technically one can move $P_{\mathbf{k}}(t_1)$ outside the integral in Eq.(9).

To obtain an additional insight into our equations we turn to the special case of static screening, which can be evaluated by integrating over t_1 in Eq.(9) by parts. Thus by using the out of integral term which is proportional to ω^{-1} we obtain

$$\int_{-\infty}^{\infty}\mathrm{Im}\{\frac{\varepsilon_\infty}{\varepsilon^*(\kappa,\omega)}\}[n(\omega)+1]\frac{d\omega}{\omega} = \int_0^{\infty}\mathrm{Im}\{\frac{\varepsilon_\infty}{\varepsilon^*(\kappa,\omega)}\}\frac{d\omega}{\omega} = \frac{\pi}{2}(1-\frac{\varepsilon_\infty}{\varepsilon(\kappa,0)}). \quad (12)$$

The result of such a calculation is

$$\begin{aligned}I_{\mathbf{k}}(t) &= P_{\mathbf{k}}(t)\sum_\kappa v_\kappa(1-\frac{\varepsilon_\infty}{\varepsilon(\kappa,0)})(f^e_{\mathbf{k}-\kappa}(t)+f^h_{\mathbf{k}-\kappa}(t)-1) -\\ &\sum_\kappa v_\kappa P_{\mathbf{k}-\kappa}(t)(1-\frac{\varepsilon_\infty}{\varepsilon(\kappa,0)})(f^e_{\mathbf{k}}(t)+f^h_{\mathbf{k}}(t)-1). \quad (13)\end{aligned}$$

In this approximation one can calculates the energy and field renormalization contributions taking into account the static screening of interaction potential. Dynamical screening is represented by the function $I_{\mathbf{k}}(t)$ Eq. (9).

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If we consider our results in the high density EHP limit, we can neglect the excitonic effect and obtain

$$(i\frac{\partial}{\partial t}-\omega_k)P_{\mathbf{k}}(t) = -\frac{1}{\hbar}E(t)d^{eh}(1-f^e_{\mathbf{k}}(t)-f^h_{\mathbf{k}}(t))+\frac{i}{\pi\hbar}\int_0^t dt_1\sum_\kappa v_\kappa\int_{-\infty}^{\infty}d\omega\,\mathrm{Im}\{\frac{\varepsilon_\infty}{\varepsilon^*(\kappa,\omega)}\}$$
$$e^{-i\omega mega(t-t_1)}[\varphi^e_{\mathbf{k}-\kappa}(t)G_{\mathbf{k}-\kappa,\mathbf{k}}(t-t_1)+\varphi^h_{\mathbf{k}-\kappa}(t)G_{\mathbf{k},\mathbf{k}-\kappa}(t-t_1)]P_{\mathbf{k}}(t_1). \quad (14)$$

A test of our result (14) is to find the steady state solution. To investigate the steady state we let $t\to\infty$ and express $P_{\mathbf{k}}(t)$ by its Fourier transform

$$P_{\mathbf{k}}(t)=\int_{-\infty}^{\infty}e^{-i\Omega t}P_{\mathbf{k}}(\Omega)d\Omega$$

If we insert this expression for the polarization $P_{\mathbf{k}}(t)$ into Eq.(13) and consider low electromegnetic field intensities we obtain

$$P_{\mathbf{k}}(t) = -E(t)d^{eh}(1-f^e_{\mathbf{k}}(t)-f^h_{\mathbf{k}}(t))\{\hbar(\omega-\omega_k)+\frac{1}{\pi}\sum_\kappa v_\kappa\int_{-\infty}^{\infty}\mathrm{Im}\{\frac{\varepsilon_\infty}{\varepsilon^*(\kappa,\Omega)}\}$$
$$\{\frac{n(\Omega)+1-f^h_{\mathbf{k}-\kappa}}{\Omega-\omega+\frac{1}{\hbar}(H_{\mathbf{k}}+E_{\mathbf{k}-\kappa})-i\gamma}+\frac{n(\Omega)+1-f^e_{\mathbf{k}-\kappa}}{\Omega-\omega+\frac{1}{\hbar}(E_{\mathbf{k}}+H_{\mathbf{k}-\kappa})-i\gamma}\}^{-1}d\Omega. \quad (15)$$

where $\hbar\omega_k$ is the exchange renormalized energy. The expression for polarization $P_{\mathbf{k}}(t)$ given by the Eq.(15) is in accordance with that obtained by Green's function method [8,17]. In this approximation only consecutive absorption/emission of more than one plasmon can be taken into account [8]. On the other hand Eq. (14) can be integrated if we use the quasi-particle picture representing by Eq. (5). If we rewrite integral over $t_1$in Eq.(14) as

$$\int_0^t dt_1\int_0^{t_1}dt_2\exp[-iw(t-t_1)]E(t_2)G_{\mathbf{k},\mathbf{k}}(t_1-t_2) = \int_0^t dt_1\int_{t_1}^t dt_2\exp[-iw(t-t_2)]E(t_1)$$
$$G_{\mathbf{k},\mathbf{k}}(t-t_1)G^*_{\mathbf{k},\mathbf{k}}(t-t_2)$$

we obtain for the Green's function the following expression

$$G_{\mathbf{k},\mathbf{k}}(t-t_1) = \exp[-i\omega_k(t-t_1)-$$
$$\frac{1}{\pi\hbar}\sum_\kappa v_\kappa\int_{-\infty}^{\infty}\mathrm{Im}\{\frac{\varepsilon_\infty}{\varepsilon^*(\kappa,\omega)}\}[(n(\omega)+1-f^e_{\mathbf{k}-\kappa})\Phi_{\mathbf{k}}(\omega-\omega^e_{\mathbf{k},\mathbf{k}-\kappa})+$$
$$(n_\omega+1-f^h_{\mathbf{k}-\kappa})\Phi_{\mathbf{k}}(\omega-\omega^h_{\mathbf{k},\mathbf{k}-\kappa})]d\omega]. \quad (16)$$

where

$$\Phi(\Omega)=\frac{-it}{\Omega}+\frac{1}{\Omega^2}[1-\exp(-i\Omega t)].$$

This result for $G_{\mathbf{k},\mathbf{k}}(t-t_1)$ differs from that evaluated by means of the cumulant expansion approximation [16]. In Eq.(16), the dynamical screening of the exchange contributions is taken into account. Eq (16)and Eq(5) have to be solved selfconsistently. Using results obtained, we can calculate the gain G_ω and the rate of spontaneous emission r_ω at arbitrary EHP concentration and temperature.

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According to Eq. (16) the frequencies of quasi-particles (plasmons, phonons) which are involved into optical transitions have to be determined from the equation $\varepsilon(\kappa,\omega)=0$. The dielectric function $\varepsilon(\kappa,\omega)$ in the random phase approximation [18] for cubic crystals in an elementary cell of which there is two atoms can be represented by the next expression

$$\varepsilon(\kappa,\omega)=\varepsilon_\infty\frac{\omega_0^2-(\omega+i\gamma)^2}{\omega_{TO}^2-(\omega+i\gamma)^2}-\frac{4\pi e^2}{\kappa^2}\{Q_e(\kappa,\omega)+Q_h(\kappa,\omega)\}, \tag{17}$$

where ω_0 is the LO-phonon frequency, $\omega_{TO}^2=\omega_0^2\varepsilon_\infty/\varepsilon_0$ is determined via the static permittivity ε_0, and

$$Q_{e,h}(\kappa,\omega)=\frac{2}{\hbar V}\sum_{\mathbf{k}}\frac{f_{\mathbf{k}}^{(e,h)}-f_{\mathbf{k}-\kappa}^{(e,h)}}{\omega+\omega_{\mathbf{k}-\kappa}^{(e,h)}+i\gamma},\gamma\to 0. \tag{18}$$

The valence electrons, the polar lattice vibrations and the EHP give additive contributions to the total dielectric function (17). Using the long-wave range approximation ($\kappa\to 0$) for $\varepsilon(\kappa,\omega)$, we obtain

$$\begin{aligned}\frac{2}{\pi}\mathrm{Im}\{\frac{\varepsilon_\infty}{\varepsilon^*(0,\omega)}\} &= c_+[\delta(\omega-\omega_+)-\delta(\omega+\omega_+)]+c_-[\delta(\omega-\omega_-)-\delta(\omega+\omega_-)], \\ c_+ &= \frac{\omega_+^2-\omega_{TO}^2}{\omega_+^2-\omega_-^2}\omega_+c_-=\frac{\omega_{TO}^2-\omega_-^2}{\omega_+^2-\omega_-^2}\omega_-.\end{aligned} \tag{19}$$

Here, $\omega_\pm$ are the eigen frequencies for longitudinal waves of the system, which are given by the zeros of the dielectric function $\varepsilon(0,\omega)$

$$\omega_\pm^2=\frac{1}{2}\{\omega_0^2+\omega_{p\infty}^2\pm[(\omega_0^2+\omega_{p\infty}^2)^2-4\omega_{p\infty}^2\omega_{TO}^2]^{1/2}\}. \tag{20}$$

The high plasmon frequency $\omega_{p\infty}$ and the plasmon frequency ω_p are

$$\omega_{p\infty}=\omega_p(\frac{\varepsilon_0}{\varepsilon_\infty})^{1/2},\omega_p=(\frac{4\pi Ne^2}{\varepsilon_0 m_r})^{1/2}. \tag{21}$$

The plasmon-phonon coupling is not strong at considerably low EHP concentration ($\omega_p<<\omega_0$), thus $\omega_+\to\omega_0$ and $\omega_-\to\omega_p$. In the limit of high electron concentrations the LO-phonon mode vibrates at the TO frequency ω_{TO} because the charge carriers have shielded out the polarization of the lattice vibrations and $\omega_-\to\omega_{TO}$, but $\omega_+\to\omega_{p\infty}$. If we insert the imaginary part of inverse dielectric function (19) into Eq.(16) we can include into the consideration the $e-h$ pairs interaction with mixed modes $\omega_\pm$ at arbitrary EHP concentration.

It can be shown, that the ground state exciton interaction with plasmons is weak due to the smallness of the Bohr radius compared to the Debye screening radius λ_D. Using obtained formulas we restrict values of plasma concentration under consideration to an interval in which the EHP concentration is sufficient enough to create Mott transition from exciton gas to EHP. The value κ_c up to which Landau damping can be neglected depends on the EHP degeneracy. For nondegenerate EHP in the limit of high temperatures $\kappa_c=1/\lambda_D$ and for degenerate EHP $\kappa_c=\kappa_{FT}$. At arbitrary EHP concentration we have

$$\kappa_c=(2m_e)^{3/2}e^2(k_0T_e)^{1/2}\hbar^{-3}\varepsilon_0^{-1}(I_e+(m_h/m_e)^{3/2}I_h). \tag{22}$$

Here, we have introduced the next designations

$$I_{e,h}=\int_0^\infty[\exp(x^2-\mu_{e,h}/k_0T_e)+1]^{-1}dx.$$

In the short-wave range limit for $\varepsilon(\kappa,\omega)$ we can neglect the EHP contributions $Q_{e,h}(\kappa,\omega)$ to the total dielectric function (17). Thus, the zero of $\varepsilon(\kappa,\omega)$ is at $\omega=\omega_0$ and $\kappa>>\kappa_c$. Hence, electron - hole pairs coupling to the

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LO-phonons have to be taken into account not only at low EHP concentrations when $\omega_+ \to \omega_0$. Short-wave length LO-phonons are coupled to the electrons and holes if $(m_{e,h}\omega_0/\hbar)^{1/2} > \kappa_c$. We assume that the inequality $m_h > m_e$ is fulfiled. Thus the emission and absorption of photon and several LO-phonons having $\kappa >> \kappa_c$ must to be included into consideration.

Using results obtained, we can get the next expression for the absorption coefficient $\alpha(\omega)$ or for the gain $G(\omega) = -\alpha(\omega)$

$$\alpha(\omega) = Ae^{\{-N_- \coth(\frac{\beta_-}{2})-N_+\}}\frac{\omega_0}{\omega}\sum_{j=-\infty}^{\infty} e^{j\frac{\beta_-}{2}} I_j(z)(g(\omega_j) + g_+ + g_{LO}). \tag{23}$$

The next designations are used here

$$g_+ = N_+ n(\omega_+) g(\omega_j + \frac{\omega_+}{\omega_0}) + \frac{1}{2}N_+^2 n(\omega_+)^2 g(\omega_j + 2\frac{\omega_+}{\omega_0}) + \frac{1}{6}N_+^3 n(\omega_+)^3 g(\omega_j + 3\frac{\omega_+}{\omega_0}),$$

$$g_{LO} = N_{LO} g(\omega_j + 1) + \frac{1}{2}N_{LO}^2 g(\omega_j + 2) + \frac{1}{6}N_{LO}^3 g(\omega_j + 3),$$

$$\omega_j = \tilde{\omega} - j\frac{\omega_-}{\omega_0}, \tilde{\omega} = \frac{\omega - \tilde{\omega}_g}{\omega_0}, \tilde{\omega}_g = \omega_g - N_-\omega_- - N_+\omega_+ - \alpha_F\omega_0\sqrt{\frac{m_h}{m_e}},$$

and

$$N_\pm = \frac{2e^2\kappa_c c_\pm}{\pi\hbar\omega_\pm^2\varepsilon_\infty}, N_{LO} = \frac{1}{2}\alpha_F n(\omega_0)\sqrt{\frac{m_h}{m_e}}, z = N_-/sh(\frac{\beta_-}{2}),$$

$$\beta_- = \hbar\omega_-/k_0T_e, g(\omega_j) = C(\omega_j)\sqrt{\omega_j}\{1 - f_e(\omega_j) - f_h(\omega_j)\}.$$

In the last expression $C(\omega_j)$ is determined by the following formula with $\omega_j = \frac{\hbar k^2}{2m_r\omega_0}$:

$$\begin{aligned} C(\mathbf{k}) &= q(\mathbf{k})e^{q(\mathbf{k})}/\mathrm{sh}q(\mathbf{k}) \\ q(\mathbf{k}) &= 2/ka_B \arctan(2\lambda_D k). \end{aligned}$$

α_F is the Frohlich coupling constant for electrons.

Expression for the spontaneous emission rate $r(\omega)$ can be write by a similar way:

$$r(\omega) = (\frac{n\omega}{\pi c})^2 Ae^{\{-N_- \coth(\frac{\beta_-}{2})-N_+\}}\frac{\omega_0}{\omega}\sum_{j=-\infty}^{\infty} e^{j\frac{\beta_-}{2}} I_j(z)(g_1(\omega_j) + g_{1,+} + g_{1,LO}). \tag{24}$$

Here

$$\begin{aligned} g_{1,+} &= N_+[n(\omega_+)+1]g_1(\omega_j + \frac{\omega_+}{\omega_0}) + \frac{1}{2}N_+^2[n(\omega_+)+1]^2 g_1(\omega_j + 2\frac{\omega_+}{\omega_0}) + \\ &\quad \frac{1}{6}N_+^3[n(\omega_+)+1]^3\ g_1(\omega_j + 3\frac{\omega_+}{\omega_0}), \end{aligned}$$

$$g_{1,LO} = N_{LO1} g_1(\omega_j + 1) + \frac{1}{2}N_{LO1}^2 * g_1(\omega_j + 2) + \frac{1}{6}N_{LO1}^3 g_1(\omega_j + 3)\},$$

$$\omega_j = \tilde{\omega} + j\frac{\omega_-}{\omega_0}, g_1(\omega_j) = C(\omega_j)\sqrt{\omega_j} * f_e(\omega_j) * f_h(\omega_j),$$

and

$$N_{LO1} = \frac{1}{2}\alpha_F[n(\omega_0)+1]\sqrt{\frac{m_h}{m_e}}.$$

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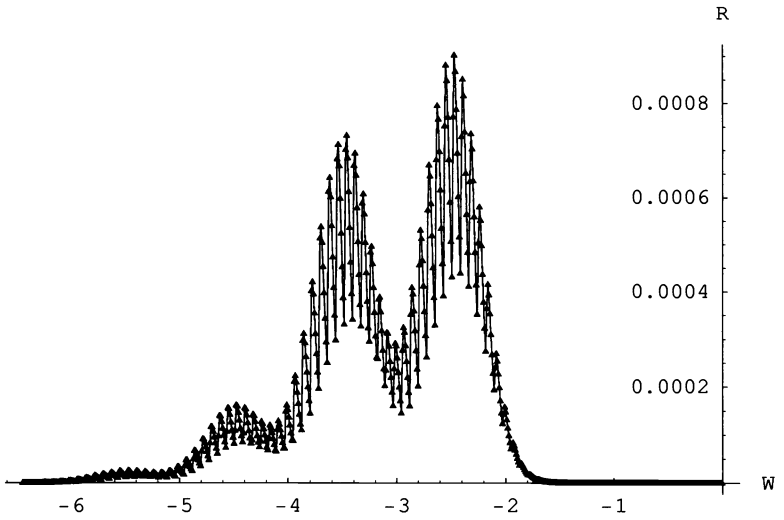


Figure 1. The spontaneous spectral function $R = r(\omega)/B$ versus the normalized photon frequency $W = (\hbar\omega - E_g)/\hbar\omega_0$ calculated using the expression (24). $B = A(\frac{n}{\pi c})^2\omega_0\omega_g$. $\hbar\omega_0 = 31meV$, $E_g = 2.7eV$, $KT = 1.5meV$, $N = 5$ $10^{15}cm^{-3}$, $m_e = 0.17m_0$, $m_h = 0.7m_0$, $\varepsilon_\infty = 5.9$, $\varepsilon_0 = 8.66$.

Three different sums enter into expressions for $\alpha(\omega)$ and $r(\omega)$. They describe a contribution into the spontaneous radiation or absorption of different processes. Thus, the first sum including $g_1(\omega_j)$ accounts multi-quantum transitions in which photon and mixed phonon-plasmons of frequency ω_- are involved. The sum with $g_{1,LO}$ accounts emission of photon, mixed phonon-plasmons and one, two and three short wave-lengths LO phonons. And at last, the sum $g_{1,+}$ includes mixed phonon-plasmons of frequency ω_+ replicas.

Numerical calculations have been carried out for two sets of parameters typical for GaAs and ZnSe (figure 1-5). These sets are different by a value of the Frohlich constant α_F and lattice parameters E_g , $m_{e,h}$. As a result, LO-phonon replicas in the spontaneous emission spectrum at low temperatures and carrier concentrations are more pronounced for ZnSe ($\alpha_F \approx 0.5$, figure 1).

The main maximum of intensity is in the domain of phononless band. With increasing the concentration (figure 2a) only two maximum are left and a relation between their values is changed that indicates the more strong increasing of the LO phonon satellite contribution into radiation. More careful analysis of the contribution of different sums shows that this increasing is connected with increasing the second and the third sums (figure 2 b,c). The spectrum has a well developed thin structure connected with MP transitions.

It is worth noting that the thin MP structure can be smooth by different factors. First, because of the Landau damping. To reduce the collision damping one can carry out the experiment at crystal temperature that corresponds to the maximum value of the mobility. Second, different recombined e-h pairs are in the places in the crystal in which plasma concentration is different one. Distribution of electrons and holes with distance from the excited crystal surface leads to the inhomogeneous broadening due to the dependence of the plasmon frequency on plasma concentration. However, the Landau damping and dispersion of plasmons are not taken into account in the present work.

At a higher temperature, the MP structure as well as LO phonon replicas become less pronounced due to the broadening of the Maxwell distribution with temperature. At low carrier concentration the first LO-phonon satellite forms only a shoulder at the red wave-length wing of the emission band (figure 3a). But at high plasma concentration maximum of intensity is in the region of the first LO-phonon replica and phononless band forms the shoulder at the short wave-lengths wing (figure 3b).

For parameters of GaAs the spectrum line given by the first sum in Eq.(24) is considerably wide even at low temperatures (figure 4). Moreover the constant $\alpha_F \approx 0.12$ is considerably small. So, the main contribution at low concentration is provided by the first sum and it determines the maximum of intensity. However, with increasing N

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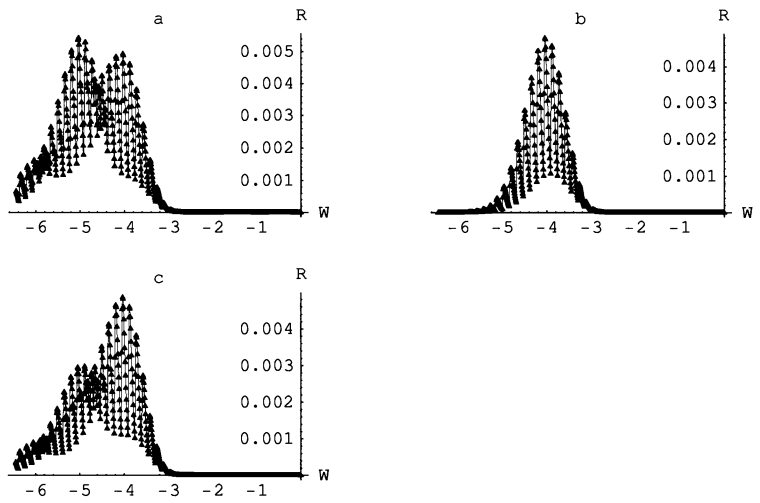


Figure 2. The spontaneous spectral function R versus the normalized photon frequency W calculated using the expression (24) (a), taking into account only the first sum in the expression (24) (b) and the first and the second sums. $N = 2\ 10^{16} cm^{-3}$, the rest parameters and disignations are as in figure 1.

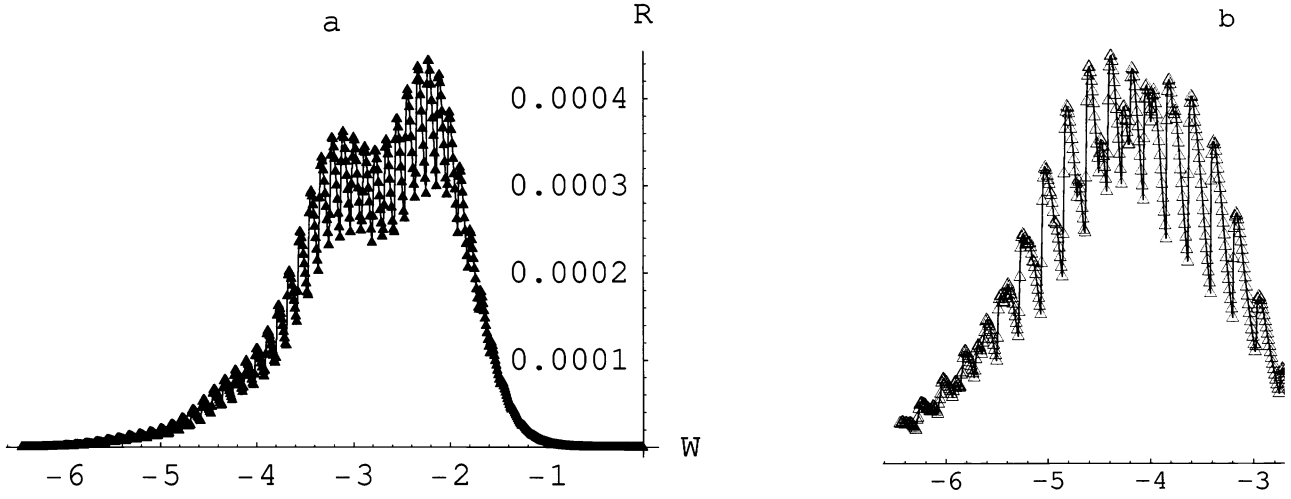


Figure 3. The spontaneous spectral function R versus the normalized photon frequency W calculated using the expression (24) (a), taking into account only the first sum in the expression (24) (b) and the first and the second sums. $KT = 4meV$, $N = 10^{16}$ (a) and $4\ 10^{16} cm^{-3}$ (b), the rest parameters and designations are as in figure 1.

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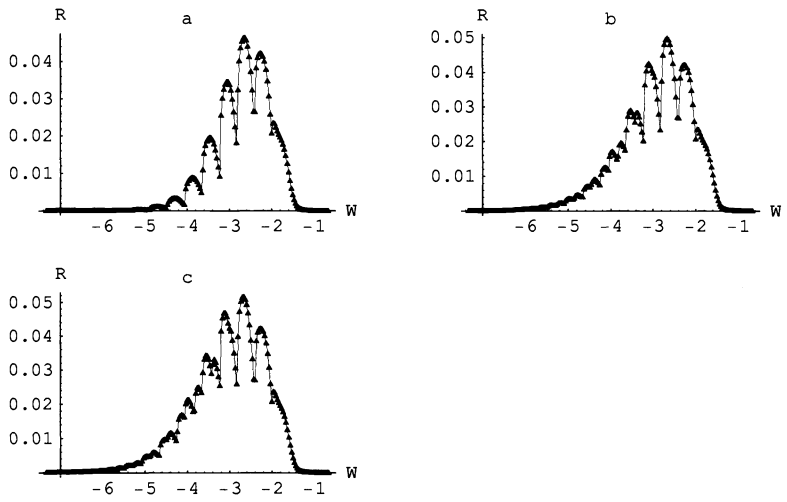


Figure 4. The spontaneous spectral function R versus the normalized photon frequency W calculated with account only for the first sum in the expression (24) (a), the first and the second sums (b) and taking into account also the third sum (c). The designations are as in figure 1. $\hbar\omega_0 = 3meV$, $E_g = 1.42eV$, $KT = 2meV$, $N = 10^{17}cm^{-3}$, $m_e = 0.072m_0$, $m_h = 0.68m_0$, $\varepsilon_\infty = 11.1$, $\varepsilon_0 = 13.13$.

the second sum strong rises in a wide spectral domain (figure 5). Its maximum corresponds to the first LO satellite. As a result the line maximum is shifted to the red side again. The competition between different transitions is not so evident as in the case of ZnSe. Phononless band forms a slightly detectable shoulder at the short wave-lengths wing at considerably high concentration $N > 10^{18}cm^{-3}$. At high EHP concentrations the coupling with long wave-length excitations ω_+ and ω_- and the interaction with short wave-lengths LO-phonons forms the main maximum of radiation in the region of the first LO-phonon satellite. These features of the spontaneous emission spectra are in accordance with the experimental results [19].

It was shown [17] that the bound state levels (exciton energies) remain practically constant whereas the continuum edge (band gap) shifts to lower energies with increasing the EHP density in accordance with experimental observations. Thus the transition from excitonic to free-carrier EHP behavior (the Mott transition) occurs at EHP concentration which can be calculated from the equation $\hbar\delta\omega_g = Ry$. This equation manifests the condition that excitonic level is just at the conduction band edge. At high temperature for nondegenerated EHP it gives $a_B/\lambda_D = 0.5$ instead of the equation $a_B/\lambda_D = 1.19$ [20], and for degenerated EHP $a_B k_F = \frac{\pi}{8}$. Using values of parameters GaN: $m_e = 0.2m_0$, $m_h = 0.76m_0, \varepsilon_0 = 9.85$ at $T = 77K$ one obtains the Mott density in this material $N_{Mott} = 3.9\ 10^{16}cm^{-3} and$ at $T = 0K$ $N_{Mott} = 5.84\ 10^{16}cm^{-3}$. At $N = 10^{18}cm^{-3}$ the plasmon energy is equal to $\hbar\omega_p = 28.8meV$. Using Eq.(21) one gets

$\hbar\ \omega_+ = 95,1meV$, $\hbar\ \omega_- = 27.5meV$. Thus at the EHP concentrations $N = 10^{17} - 10^{18}cm^{-3}$ the mixing of the plasmon and LO-phonon modes can be neglected. On the other hand, the EHP concentration is high enough that the Mott transition occurs. Photoluminescence spectra are then dominated by EHP emission consisting of individual peaks approximately spaced by the LO-phonon energy of 91 meV one from other. Amplitude ratio between the phononless and the first LO-phonon replica is decreased with EHP concentration increasing . All these features of the luminescence spectra are in accordance with that obtained in the experiments with GaN samples at the bath temperature of 30 K at different incident power densities [19]. At high excitation density the free-to free transitions within the EHP was observed. Several structures, almost periodic, appear below the phonon-less peak and was identified as phonon replicas[19]. Besides, the thin structure attributed to the low frequency modes (ω_-) is presented at the luminescence spectra at high level of excitations.

Another problem is connected with determination of the half-width of emission band . For free-carrier emission mechanism the half-width have to be equal to $1.8kT$ (about $30meV$ at $200K$). Experimental data and this simple

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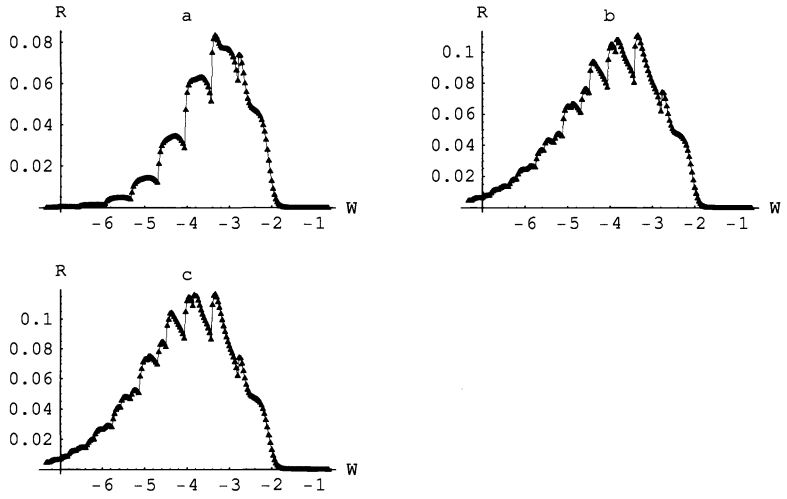


Figure 5. The spontaneous spectral function R versus the normalized photon frequency W calculated with account only for the first sum in the expression (24) (a), the first and the second sums (b) and taking into account also the third sum (c). $N = 2.5\ 10^{17} cm^{-3}$, the rest parameters and designations are as in figure 4.

theory prediction do not coincide. Basing on our results we can conclude that multi-plasmon processes determine the shape of the emission band.

Strong increasing with N of the first LO-phonon replica contribution into emission of GaN:Mg at bound-to-bound (DAP) transitions was observed in the paper [21] (see Fig.9).

4. CONCLUSIONS

In this paper we have treated the polarization in a crystal under the influence of a classical alternative electric field at the level of microscopic theory. Beyond Hartree-Fock approximation the kinetic equation derived for polarization and carrier occupation probabilities contain correlation contributions describing multi-quantum optical transitions involving photon and a several mixed phonon-plasmons of frequencies ω_+ , ω_-.

The approximation is based on finding the collision integral which contains all terms proportional to v_κ^2 with using of the fluctuation-dissipation theorem. To account for static screening effects from the correlations authors of works [1-6] replace the unscreened Coulomb potential v_κ by the screened one. Green's functions approach presents a consistent way to include strong higher-order Coulomb correlations within the coupled photon and carrier system [7-9]. We believe that using of the fluctuation-dissipation theorem is the simplest way to take into consideration the infinitive series in degrees of v_κ of the perturbation theory. It is shown that the using of fluctuation-dissipation theorem is the simplest way to take into consideration the high-order Coulomb correlations and avoid the divergency.

The numerical results obtained are physically reasonable in view of existing experimental results .

One might be able to improve the accuracy of our result by including another higher-order correlation contributions into the consideration. For example, the six-particle correlation functions, representing six - wave mixing by exciton to two exciton transitions and light quantization effects like non classical statistics, squeezing of the light fluctuations or entanglement between light and matter excitations [1].

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REFERENCES

1. S.W. Koch, M.Kira, T.Meier. Journal of Optics B, 2001(to be published).
2. M. Bonitz, J.W. Dufty, Chang Sub Kim. Phys. Stat. Sol. (b), **206**, 181 (1998).
3. H. Haug, S.W. Koch. Quantum *Theory of the Optical and Electronic Properties of Semiconductors, 3rd ed.* (Word Scientific, Singapore, 1994).
4. W.W. Chow, S.W. Koch, M. Sargent III. *Semiconductor-Laser Physics* (Springer-Verlag, Berlin, 1994).
5. A. Girndt, S.W. Koch, W.W. Chow. Appl. Phys. A **66**, 1 (1998).
6. W.W. Chow, A. Knorr, S. Hughes, A. Girndt, S.W. Koch. IEEE J. of Selected Topics in Quant. Electron., **3**, 136 (1997).
7. U.Moldzio, G.Manzke, K.Henneberger. Phys. Stat. Sol. (b), **206**, 265 (1998).
8. R. Schepe, T. Schmelau, D. Tamme, K. Henneberger. Phys. Stat. Sol. (b), **206**, 273 (1998).
9. M.F.Pereira,Jr. K.Hennenberger. Phys. Stat. Sol. (b), **206**, 477 (1998).
10. A.A. Klyukanov. Fizika Tverdogo Tela, **29**, 1529 (1987).
11. V.S. Vavilov, A.A. Klyukanov, K.D. Sushkevich, M.V. Chukichev, R.R. Resvanov, S.K. Sushkevich. Fizika Tverdogo Tela, **39**, 1526 (1997).
12. A.A. Klyukanov, E.A. Senokosov, L.E. Chibotaru. Phys.Stat. Sol. (b), **155**,295 (1989).
13. V.S. Vavilov, A.A. Klyukanov, K.D. Sushkevich, M.V. Chukichev, R.R. Resvanov. Fizika Tverdogo Tela, **41**,1176 (1999).
14. V.S. Vavilov, A.A. Klyukanov, M.V. Chukichev, O.M. Shapoval. A.Z. Awawdeh, R.R. Resvanov. Fizika i Tekhnika Poluprovodnikov, **28**, 2113 (1994).
15. V.S. Vavilov, A.A. Klyukanov, K.D. Sushkevich, M.V.Chukichev, A.Z. Awawdeh, R.R. Resvanov. Fizika Tverdogo Tela, **37**, 312 (1995).
16. A.A. Klyukanov, N.A.Loiko, I.Babushkin. Laser Physics, 11, No 3 (2001).
17. R. Zimmerman, K. Kilimann, W.D.Krafet, D.Kremp, G. Ropke. Phys. Stat. Sol(b), **90**, 175 (1978).
18. P.M. Platsman, P.A. Wolff. *Waves and interactions in solid state plasmas* (Academic press, New-York and London, 1973)
19. F.Binet, J.Y.Duboz, J.Off,F.Scholz. Phys.Rev.**60**, 4715,(1999).
20. R.C,Casella, J.Appl.Phys.**34**,1703,(1963).
21. P.H.Lim,B.Schineller,O.Schon,K.Heime,M.Heuken.J.Crystal Growth,**205**,1,(1999).

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