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Nonmonotonic decay of nonequilibrium polariton condensate in direct-gap semiconductors

Igor V. Beloussov and Vladimir V. Frolov
Institute of Applied Physics, Academy of Sciences of Moldavia, Academy Street 5, 277028 Kishinev, Moldavia

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Time evolution of a nonequilibrium polariton condensate has been studied in the framework of a microscopic approach. It has been shown that due to polariton-polariton scattering a significant condensate depletion takes place in a comparatively short time interval. The condensate decay occurs in the form of multiple echo signals. Distribution-function dynamics of noncondensate polaritons have been investigated. It has been shown that at the initial stage of evolution the distribution function has the form of a bell. Then oscillations arise in the contour of the distribution function, which further transform into small chaotic ripples. The appearance of a short-wavelength wing of the distribution function has been demonstrated. We have pointed out the enhancement and then partial extinction of the sharp extra peak arising within the time interval characterized by small values of polariton condensate density and its relatively slow changes. [S0163-1829(96)00127-0]

I. INTRODUCTION

In a great number of works concerning spatial and time evolution of laser radiation in resonant media the classical description is used when the photon and material fields are considered as interacting macroscopically occupied coherent modes (condensates). When a quasiparticle quits a condensate mode, that is usually taken into account by introduction of phenomenological damping constants. In this case it is assumed that (1) the condensate decay is exponential and (2) the condensate lifetime is considerably longer than the duration of the coherent nonlinear process being investigated.

From our point of view it is more consistent to use a microscopic approach (see, e.g., Refs. 1–3), where the dynamics of interacting condensate modes and quantum fluctuations appearing in a system as a result of real scattering processes of quasiparticles are described on equal footing. In other words, one should take into account (1) the interaction between condensates, (2) excitation of noncondensate particles, and (3) the backward influence of noncondensate particles on the condensates.

The microscopic approach is used in this work to describe the time evolution of the system of initially coherent high-density excitons and photons.

It is known[4] that coherent electromagnetic radiation resonant to the isolated exciton energy level excites in the crystal the coherent polariton wave with the wave vector $\mathbf{k}_0 \neq 0$ — the nonequilibrium polariton condensate. Different scattering processes accompanying its propagation lead to the loss of initial coherence of the polariton wave, complete or partial depletion of condensate, excitation of polaritons characterized by some statistics with wave vector $\mathbf{k} \neq \mathbf{k}_0$, and other phenomena.

At sufficiently high excitation energies the processes of polariton scattering due to exciton-exciton collisions prevail. This scattering mechanism is of considerable interest due to the recent experimental investigations[5] and many interesting physical results obtained in theoretical study of dynamic and kinetic processes in the system of interacting polaritons.

According to Refs. 6 and 7 exciton-exciton scattering is of significant importance in the situation where coherent polaritons are excited in a certain spectral region where energy and momentum conservation laws allow real processes of two-quantum excitation of polaritons from the condensate. These processes lead to instability of the completely condensed state of the polariton system. Existence of this spectral region situated around the isolated exciton resonance is due to the peculiarities of the polariton dispersion law.

In Refs. 6 and 7 the energy spectrum of noncondensate polaritons, arising as a result of decay of coherent polariton wave, is studied. According to Refs. 6 and 7 in some regions of **k** space the energy spectrum does not exist.

It should be mentioned that investigations performed in Refs. 6 and 7 are based on the model formally analogous to that used by Bogoliubov in Ref. 8 to study the equilibrium system of weakly nonideal Bose gas. In the nonequilibrium situation considered in Refs. 6 and 7, when decay of the polariton condensate and excitation of noncondensate polaritons take place, this model is adequate to the real situation only at the initial stage of the condensate decay when the number of polaritons in the condensate is still much greater than the total number of noncondensate polaritons. But this stage is essentially *nonstationary* and the occurrence of condensate instability provides evidence for that. The study of the energy spectrum implies the determination of the *steady states* of the system.[9] That is why the results of Refs. 6 and 7, concerning the energy spectrum of the system, based on the above-mentioned model, in our opinion cannot be regarded as well grounded. This remark refers to the relatively recent works[10,11] as well.

Because of essential nonstationarity of the processes in the system methods of nonequilibrium mechanics should be used to describe it adequately. Derivation of the equations that describe kinetics of the polariton condensate decay and excitation of quantum fluctuations has some specific features owing to degeneracy in the system. As the energy and resulting wave vector of two non-condensate polaritons can be equal to the energy and wave vector of two condensate polaritons, respectively, there is degeneracy of two-particle states. Moreover, the presence of the condensate in the system also leads to degeneracy due to its macroscopic amplitude.[12]

The correct description of the system with degeneracy requires the introduction of abnormal distribution functions[13]

$$\Psi_{\mathbf{k}_0}=\langle\Phi_{\mathbf{k}_0}\rangle$$

and

$$F_{\mathbf{k}}=\langle\Phi_{\mathbf{k}}\Phi_{2\mathbf{k}_0-\mathbf{k}}\rangle$$

together with the normal (usual) ones

$$N_{\mathbf{k}}=\langle\Phi^{\dagger}_{\mathbf{k}}\Phi_{\mathbf{k}}\rangle.$$

Here $\Phi^{\dagger}_{\mathbf{k}}$ $(\Phi_{\mathbf{k}})$ are Bose operators of creation (annihilation) of a polariton on the lower branch with the wave vector **k**. The appearance of abnormal averages indicates a breaking of the selection rules connected with the gauge invariance of the system.[13–15] In this problem the breaking of gauge symmetry arises as a result of the noninvariant initial condition due to the assumption that there is a condensate in the system at the initial moment $t=t_0$.

An earlier attempt to obtain kinetic equations for polaritons excited in semiconductors by the external classic field was made in Refs. 16 and 17. However, in Refs. 16 and 17 degeneracy of two-particle states was not taken into account and the abnormal distribution functions $F_{\mathbf{k}}$ were not introduced. So one should expect the equations obtained in Refs. 16 and 17 to possess unphysical singularity.

Kinetic equations describing evolution of partially coherent polaritons that take into account the degeneracy were obtained in Refs. 18 and 19 using the nonequilibrium Green's-function technique,[20,21] presented by the authors in terms of functionals. They coincide with the equations obtained in Ref. 22 using the method of nonequilibrium statistical operator,[23] and do not possess unphysical singularities.

According to Refs. 19 and 22 the kinetics of partially coherent polaritons is described in the Born approximation by the closed set of nonlinear integrodifferential equations for the coherent part of polariton field $\Psi_{\mathbf{k}_0}$ and the normal

$$n_{\mathbf{k}}=N_{\mathbf{k}}-\delta_{\mathbf{k},\mathbf{k}_0}|\Psi_{\mathbf{k}_0}|^2$$

and abnormal

$$f_{\mathbf{k}}=F_{\mathbf{k}}-\delta_{\mathbf{k},\mathbf{k}_0}\Psi^2_{\mathbf{k}_0}$$

distribution functions. In the absence of quantum fluctuations, described by the functions $n_{\mathbf{k}}$ and $f_{\mathbf{k}}$, the equations for them become identities, and the equation for $\Psi_{\mathbf{k}\alpha}$ (α is the number of polariton branch) coincides with that obtained in Ref. 24 for the system of interacting coherent excitons and photons. In another particular case, when $\Psi_{\mathbf{k}_0}=0$ and $f_{\mathbf{k}}=0$, the equations obtained in Refs. 19 and 22 are reduced to the usual kinetic equation for the distribution function $N_{\mathbf{k}}$ (see, e.g., Refs. 23 and 25).

The right-hand sides of kinetic equations obtained in Refs. 19 and 22 include terms linear in the constant of exciton-exciton interaction $\nu>0$ and the ones $\sim\nu^2$. Terms $\sim\nu$ correspond to the self-consistent-field approximation, which neglects the higher-order correlation functions. In this approximation the processes of two-particle excitations of polaritons from the condensate, backward processes, and transformation of a created pair of noncondensate polaritons into another pair with the same value of total momentum are taken into account. It describes the fastest processes in the system and is sufficient for study of the early (before-kinetic) stage of the system's evolution.

Terms $\sim\nu^2$ take into account scattering processes in which only one polariton belongs to the condensate. Therefore, they differ from zero only if noncondensate polaritons exist in the system. Terms $\sim\nu^2$ in comparison with terms $\sim\nu$ describe slower changes of the system characteristics and are significant only at the kinetic stage of evolution.

Our study of time evolution of the nonequilibrium polariton system is based on the self-consistent-field approximation. This approximation has shown a good performance in the theoretical study of states of the electron-hole subsystem in semiconductors that appear just after the transmission of the front of the ultrashort laser pulse.[26,2,27] The part of evolution equations obtained in our work for the stationary case are in many respects similar to the equations given in Refs. 26, 2, and 27. Nevertheless, that slight distinctions are important and lead to significant physical consequencies.

The stationary equations obtained in Refs. 26, 2, and 27 possess nontrivial solutions describing states of the electron-hole subsystem. The decay of these states occurs only due to incoherent relaxation processes. In Refs. 26, 2 and 27 it is assumed that these processes do not have time to happen during the period of pulse action. This assumption justifies the use of the self-consistent field approximation. The attempt to find the steady-state solution of similar equations in the framework of the physical problem of this paper leads to instabilities. Their physical nature has been discussed above. As noted, the appearance of the instability points leads to essential nonstationarity of the processes in the system and requires the return to the starting (nonstationary) equations.

Moreover, the excitation of the great number of noncondensate modes can lead to significant depletion of the condensate. So to take into account this phenomenon it is necessary, along with equations similar to equations in Refs. 26, 2, and 27 to consider an additional equation for the condensate wave function $\Psi_{\mathbf{k}_0}$. Note that in Refs. 26, 2, and 27 backward influence of the electron-hole subsystem on the laser radiation field was not taken into account.

Note also Refs. 28–32 where the self-consistent-field approximation allowed us to take into account the biexciton complex structure.

II. SYSTEM HAMILTONIAN

We start from the Hamiltonian

$$\hat{H}=\hat{H}_0+\hat{H}_{int}, \tag{1}$$

$$\hat{H}_0=\sum_{\mathbf{k}}\sum_{\alpha_1,\alpha_2=1,2}(h_{\mathbf{k}})_{\alpha_1\alpha_2}\hat{\varphi}^{\dagger}_{\mathbf{k}\alpha_1}\hat{\varphi}_{\mathbf{k}\alpha_2},\quad h_{\mathbf{k}}=\hbar\omega^{\perp}_{\mathbf{k}}\left(\frac{1+\tau_3}{2}\right)+\hbar\omega_{\mathbf{k}}\left(\frac{1-\tau_3}{2}\right)+\eta_{\mathbf{k}}\tau_2, \tag{2}$$

$$\hat{H}_{\text{int}}=\frac{1}{2V}\sum_{\mathbf{k}_1,\ldots,\mathbf{k}_4}\sum_{\alpha_1,\ldots,\alpha_4=1,2}\nu_{\mathbf{k}_1-\mathbf{k}_4}\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4}\left(\frac{1+\tau_3}{2}\right)_{\alpha_1,\alpha_3}\left(\frac{1+\tau_3}{2}\right)_{\alpha_2,\alpha_4}\hat{\varphi}^{\dagger}_{\mathbf{k}_1\alpha_1}\hat{\varphi}^{\dagger}_{\mathbf{k}_2\alpha_2}\hat{\varphi}_{\mathbf{k}_3\alpha_3}\hat{\varphi}_{\mathbf{k}_4\alpha_4}. \tag{3}$$

In (1)–(3) the following notations are used. The operators $\hat{\varphi}_{\mathbf{k}\alpha}$ $(\hat{\varphi}^{\dagger}_{\mathbf{k}\alpha})$ of exciton (when $\alpha=1$) or photon (when $\alpha=2$) annihilation (creation) in the state with wave vector $\mathbf{k}$ obey Bose-type commutation relations:

$$[\hat{\varphi}_{\mathbf{k}_1\alpha_1},\hat{\varphi}^{\dagger}_{\mathbf{k}_2\alpha_2}]=\delta_{\mathbf{k}_1,\mathbf{k}_2}\delta_{\alpha_1,\alpha_2},[\hat{\varphi}_{\mathbf{k}_1\alpha_1},\hat{\varphi}_{\mathbf{k}_2\alpha_2}]=[\hat{\varphi}^{\dagger}_{\mathbf{k}_1\alpha_1},\hat{\varphi}^{\dagger}_{\mathbf{k}_2\alpha_2}]=0.$$

The Pauli matrices τ_1, τ_2, and τ_3 are taken in the standard representation.[20] The frequencies $\omega_{\mathbf{k}}$ and $\omega^{\perp}_{\mathbf{k}}$ are given by the expressions $\omega_{\mathbf{k}}=c|\mathbf{k}|\epsilon_B^{-1/2},\omega^{\perp}_{\mathbf{k}}=\omega^{\perp}+\hbar\mathbf{k}^2/2m$, where c is the vacuum velocity of light, $\hbar\omega^{\perp}$ is the exciton formation energy in the band $\hbar\omega^{\perp}_{\mathbf{k}}$, and m is its effective mass. The background dielectric function ϵ_B includes the contributions from all excitations in a crystal except the excitons of the isolated band $\hbar\omega^{\perp}_{\mathbf{k}}$. In the vicinity of the exciton resonance $\omega_{\mathbf{k}}\approx\omega^{\perp}_{\mathbf{k}}$ weak frequency dependence of ϵ_B can be neglected.

The Hamiltonian (1) describes the system of interacting dipole-active excitons and photons with transverse polarization in an infinite crystal ($V\rightarrow\infty$, V is the quantization volume) in the vicinity of an isolated exciton resonance $\omega_{\mathbf{k}}\approx\omega^{\perp}_{\mathbf{k}}$. In other words, we assume that the relations $\hbar|\omega_{\mathbf{k}}-\omega^{\perp}_{\mathbf{k}}|\ll\hbar\omega^{\perp}_{\mathbf{k}}$, $\Delta\mathcal{E}$ are imposed on the photon frequency $\hbar\omega_{\mathbf{k}}$, the energy of the exciton $\hbar\omega^{\perp}_{\mathbf{k}}$, and the minimum energy gap $\Delta\mathcal{E}$ between the exciton band $\hbar\omega^{\perp}_{\mathbf{k}}$ and any other one. Beside that we suppose that the constants of exciton-photon ($\eta_{\mathbf{k}}=\eta_{-\mathbf{k}}$) and exciton-exciton ($\nu_{\mathbf{k}}=\nu_{-\mathbf{k}}$) interactions are small enough: $0<\eta_{\mathbf{k}}/\hbar\ll\omega^{\perp}_{\mathbf{k}}$, $0<(\nu_{\mathbf{k}}/\hbar)\overline{n}\ll\omega^{\perp}_{\mathbf{k}}$ ($\overline{n}$ is an average density of excitons in the system). These assumptions allow us to retain in (2) only resonant terms and treat the exciton-exciton interaction as a small perturbation.

The quadratic part (2) of Hamiltonian (1) can be reduced to the diagonal form

$$\hat{H}_0=\sum_{\mathbf{k}}\sum_{\alpha=1,2}\hbar\Omega_{\mathbf{k}\alpha}\hat{\Phi}^{\dagger}_{\mathbf{k}\alpha}\hat{\Phi}_{\mathbf{k}\alpha} \tag{4}$$

by transition to the polariton Bose operators $\hat{\Phi}_{\mathbf{k}\alpha}$ and $\hat{\Phi}^{\dagger}_{\mathbf{k}\alpha}$ using the unitary transformation

$$\hat{\varphi}_{\mathbf{k}\alpha_1}=\sum_{\alpha_2=1,2}(U_{\mathbf{k}})_{\alpha_1\alpha_2}\hat{\Phi}_{\mathbf{k}\alpha_2},\quad U_{\mathbf{k}}=\frac{1-i\tau_1L_{\mathbf{k}}}{\sqrt{1+L^2_{\mathbf{k}}}}.$$

$L_{\mathbf{k}}$ is a function determined by the equation $\eta_{\mathbf{k}}L^2_{\mathbf{k}}+\hbar(\omega_{\mathbf{k}}-\omega^{\perp}_{\mathbf{k}})L_{\mathbf{k}}-\eta_{\mathbf{k}}=0$. In the polariton representation we have

$$\hat{H}_{\text{int}}=\frac{1}{2V}\sum_{\mathbf{k}_1,\ldots,\mathbf{k}_4}\sum_{\alpha_1,\ldots,\alpha_4=1,2}\nu_{\mathbf{k}_1-\mathbf{k}_4}\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4}\times(\mathcal{P}_{\mathbf{k}_1\mathbf{k}_3})_{\alpha_1\alpha_3}(\mathcal{P}_{\mathbf{k}_2\mathbf{k}_4})_{\alpha_2\alpha_4}\hat{\Phi}^{\dagger}_{\mathbf{k}_1\alpha_1}\hat{\Phi}^{\dagger}_{\mathbf{k}_2\alpha_2}\hat{\Phi}_{\mathbf{k}_3\alpha_3}\hat{\Phi}_{\mathbf{k}_4\alpha_4}, \tag{5}$$

where

$$\mathcal{P}_{\mathbf{k}_1\mathbf{k}_2}=\mathcal{P}^{\dagger}_{\mathbf{k}_2\mathbf{k}_1}=U^{\dagger}_{\mathbf{k}_1}\left(\frac{1+\tau_3}{2}\right)U_{\mathbf{k}_2}.$$

Hamiltonian (1) is invariant with respect to the gauge transformation $\hat{R}=\exp(i\gamma\hat{N})$ where γ is an arbitrary real parameter, $\hat{N}=\Sigma_{\mathbf{k}}\Sigma_{\alpha=1,2}\hat{\Phi}^{\dagger}_{\mathbf{k}\alpha}\hat{\Phi}_{\mathbf{k}\alpha}$ is the operator of the total number of polaritons in the system.

III. SELF-CONSISTENT FIELD APPROXIMATION

In the Heisenberg picture operators $\check{\Phi}_{\mathbf{k}\alpha}(t)$ are governed by the equation of motion

$$i\hbar\frac{d}{dt}\check{\Phi}_{\mathbf{k}\alpha}(t)=[\check{\Phi}_{\mathbf{k}\alpha}(t),\check{H}] \tag{6}$$

and average value of a dynamic quantity A is given by $\langle A\rangle_t=\text{Tr}\check{\rho}\check{A}(t)$. Here $\check{\rho}$ is the density matrix, which describes the polariton distribution at the initial moment of time $t=t_0$.

The presence of the condensate in the system implies that the coherent part of the polariton field

$$\Psi_{\mathbf{k}\alpha}(t)=\langle\Phi_{\mathbf{k}\alpha}\rangle_t \tag{7}$$

is nonzero. We write down the coherent part explicitly

$$\check{\Phi}_{\mathbf{k}\alpha}(t)=\Psi_{\mathbf{k}\alpha}(t)+\check{\chi}_{\mathbf{k}\alpha}(t). \tag{8}$$

According to definition (7), we have $\langle\check{\chi}_{\mathbf{k}\alpha}\rangle_t=0$.

Using (8) we present the Hamiltonian $\check{H}=\check{H}_0+\check{H}_{int}$ in the form $\check{H}=\check{H}_1(t)+\check{H}_2(t)$, where the operator $\check{H}_1(t)$ includes only linear and quadratic terms with respect to $\check{\chi}_{\mathbf{k}\alpha}(t)$ and $\check{\chi}^{\dagger}_{\mathbf{k}\alpha}(t)$. The operator $\check{H}_2(t)$ includes products of three and four operators $\check{\chi}_{\mathbf{k}\alpha}(t)$ and $\check{\chi}^{\dagger}_{\mathbf{k}\alpha}(t)$.

Further we shall treat $\check{H}_2(t)$ in the self-consistent field approximation. For this purpose we make the formal substitution[27]

$$\begin{aligned}
\check{\chi}^{\dagger}_{\mathbf{k}_1\alpha_1}(t)\check{\chi}^{\dagger}_{\mathbf{k}_2\alpha_2}(t)\check{\chi}_{\mathbf{k}_3\alpha_3}(t)\check{\chi}_{\mathbf{k}_4\alpha_4}(t)\rightarrow{}& \check{\chi}^{\dagger}_{\mathbf{k}_1\alpha_1}(t)\check{\chi}^{\dagger}_{\mathbf{k}_2\alpha_2}(t)\langle\chi_{\mathbf{k}_3\alpha_3}\chi_{\mathbf{k}_4\alpha_4}\rangle_t+\check{\chi}_{\mathbf{k}_3\alpha_3}(t)\check{\chi}_{\mathbf{k}_4\alpha_4}(t)\langle\chi^{\dagger}_{\mathbf{k}_1\alpha_1}\chi^{\dagger}_{\mathbf{k}_2\alpha_2}\rangle_t\\
&+\check{\chi}^{\dagger}_{\mathbf{k}_1\alpha_1}(t)\check{\chi}_{\mathbf{k}_3\alpha_3}(t)\langle\chi^{\dagger}_{\mathbf{k}_2\alpha_2}\chi_{\mathbf{k}_4\alpha_4}\rangle_t+\check{\chi}^{\dagger}_{\mathbf{k}_1\alpha_1}(t)\check{\chi}_{\mathbf{k}_4\alpha_4}(t)\langle\chi^{\dagger}_{\mathbf{k}_2\alpha_2}\chi_{\mathbf{k}_3\alpha_3}\rangle_t\\
&+\check{\chi}^{\dagger}_{\mathbf{k}_2\alpha_2}(t)\check{\chi}_{\mathbf{k}_3\alpha_3}(t)\langle\chi^{\dagger}_{\mathbf{k}_1\alpha_1}\chi_{\mathbf{k}_4\alpha_4}\rangle_t+\check{\chi}^{\dagger}_{\mathbf{k}_2\alpha_2}(t)\check{\chi}_{\mathbf{k}_4\alpha_4}(t)\langle\chi^{\dagger}_{\mathbf{k}_1\alpha_1}\chi_{\mathbf{k}_3\alpha_3}\rangle_t\,,
\end{aligned}$$

$$\check{\chi}^{\dagger}_{\mathbf{k}_2\alpha_2}(t)\check{\chi}_{\mathbf{k}_3\alpha_3}(t)\check{\chi}_{\mathbf{k}_4\alpha_4}(t)\rightarrow\check{\chi}^{\dagger}_{\mathbf{k}_2\alpha_2}(t)\langle\chi_{\mathbf{k}_3\alpha_3}\chi_{\mathbf{k}_4\alpha_4}\rangle_t+\check{\chi}_{\mathbf{k}_3\alpha_3}(t)\langle\chi^{\dagger}_{\mathbf{k}_2\alpha_2}\chi_{\mathbf{k}_4\alpha_4}\rangle_t+\check{\chi}_{\mathbf{k}_4\alpha_4}(t)\langle\chi^{\dagger}_{\mathbf{k}_2\alpha_2}\chi_{\mathbf{k}_3\alpha_3}\rangle_t\,.$$

As a result, $\check{H}_2(t)$ (and $\check{H}$) has the same operator structure as $\check{H}_1(t)$. Performing transformation from the operators $\check{\chi}_{\mathbf{k}\alpha}(t)$ back to the operators $\check{\Phi}_{\mathbf{k}\alpha}(t)$ we obtain

$$\begin{aligned}
\check{H}={}&E_0(t)+\sum_{\mathbf{k}}\sum_{\alpha=1,2}\hbar\Omega_{\mathbf{k}\alpha}\check{\Phi}^{\dagger}_{\mathbf{k}\alpha}(t)\check{\Phi}_{\mathbf{k}\alpha}(t)+\frac{1}{2V}\sum_{\mathbf{k}_1,\ldots,\mathbf{k}_4}\sum_{\alpha_1,\ldots,\alpha_4=1,2}\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4}\bigg\{[\nu_{\mathbf{k}_1-\mathbf{k}_4}(\mathcal{P}_{\mathbf{k}_1\mathbf{k}_3})_{\alpha_1\alpha_3}(\mathcal{P}_{\mathbf{k}_2\mathbf{k}_4})_{\alpha_2\alpha_4}\\
&+\nu_{\mathbf{k}_1-\mathbf{k}_3}(\mathcal{P}_{\mathbf{k}_1\mathbf{k}_4})_{\alpha_1\alpha_4}(\mathcal{P}_{\mathbf{k}_2\mathbf{k}_3})_{\alpha_2\alpha_3}]\bigg[\check{\Phi}^{\dagger}_{\mathbf{k}_1\alpha_1}(t)\check{\Phi}_{\mathbf{k}_3\alpha_3}(t)\langle\Phi^{\dagger}_{\mathbf{k}_2\alpha_2}\Phi_{\mathbf{k}_4\alpha_4}\rangle_t+\frac{1}{2}\check{\Phi}^{\dagger}_{\mathbf{k}_1\alpha_1}(t)\check{\Phi}^{\dagger}_{\mathbf{k}_2\alpha_2}(t)\langle\Phi_{\mathbf{k}_3\alpha_3}\Phi_{\mathbf{k}_4\alpha_4}\rangle_t\\
&-2\check{\Phi}^{+}_{\mathbf{k}_1\alpha_1}(t)\Psi^{*}_{\mathbf{k}_2\alpha_2}(t)\Psi_{\mathbf{k}_3\alpha_3}(t)\Psi_{\mathbf{k}_4\alpha_4}(t)\bigg]+\text{ H.c.}\bigg\},
\end{aligned}\tag{9}$$

where $E_0(t)$ is a c-number function.

With the help of Hamiltonian (9) and equations of motion (6) we obtain the set of equations, which describes time evolution of the system of partially coherent polaritons:

$$\begin{aligned}
\bigg[i\hbar\frac{d}{dt}-\hbar\Omega_{\mathbf{k}_1\alpha_1}\bigg]\Psi_{\mathbf{k}_1\alpha_1}(t)={}&\frac{1}{V}\sum_{\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4}\sum_{\alpha_2,\alpha_3,\alpha_4=1,2}\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4}[\nu_{\mathbf{k}_1-\mathbf{k}_4}(\mathcal{P}_{\mathbf{k}_1\mathbf{k}_3})_{\alpha_1\alpha_3}(\mathcal{P}_{\mathbf{k}_2\mathbf{k}_4})_{\alpha_2\alpha_4}\\
&+\nu_{\mathbf{k}_1-\mathbf{k}_3}(\mathcal{P}_{\mathbf{k}_1\mathbf{k}_4})_{\alpha_1\alpha_4}(\mathcal{P}_{\mathbf{k}_2\mathbf{k}_3})_{\alpha_2\alpha_3}]\bigg[\frac{1}{2}\Psi^{*}_{\mathbf{k}_2\alpha_2}(t)\Psi_{\mathbf{k}_3\alpha_3}(t)\Psi_{\mathbf{k}_4\alpha_4}(t)+n(\mathbf{k}_2,\alpha_2;\mathbf{k}_3,\alpha_3|t)\Psi_{\mathbf{k}_4\alpha_4}(t)\\
&+\frac{1}{2}\Psi^{*}_{\mathbf{k}_2\alpha_2}(t)f(\mathbf{k}_3,\alpha_3;\mathbf{k}_4,\alpha_4|t)\bigg];
\end{aligned}\tag{10}$$

$$\begin{aligned}
\bigg[i\hbar\frac{d}{dt}+\hbar(\Omega_{\mathbf{k}_1\alpha_1}-\Omega_{\mathbf{k}_2\alpha_2})\bigg]n(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t)={}&-\frac{1}{V}\sum_{\mathbf{k}_3,\mathbf{k}_4,\mathbf{k}_5}\sum_{\alpha_3,\alpha_4,\alpha_5=1,2}\delta_{\mathbf{k}_1+\mathbf{k}_3,\mathbf{k}_4+\mathbf{k}_5}[\nu_{\mathbf{k}_1-\mathbf{k}_5}(\mathcal{P}_{\mathbf{k}_4\mathbf{k}_1})_{\alpha_4\alpha_1}(\mathcal{P}_{\mathbf{k}_5\mathbf{k}_3})_{\alpha_5\alpha_3}\\
&+\nu_{\mathbf{k}_1-\mathbf{k}_4}(\mathcal{P}_{\mathbf{k}_5\mathbf{k}_1})_{\alpha_5\alpha_1}(\mathcal{P}_{\mathbf{k}_4\mathbf{k}_3})_{\alpha_4\alpha_3}]\bigg\{n^{*}(\mathbf{k}_2,\alpha_2;\mathbf{k}_5,\alpha_5|t)[n^{*}(\mathbf{k}_3,\alpha_3;\mathbf{k}_4,\alpha_4|t)\\
&+\Psi_{\mathbf{k}_3\alpha_3}(t)\Psi^{*}_{\mathbf{k}_4\alpha_4}(t)]+\frac{1}{2}f(\mathbf{k}_2,\alpha_2;\mathbf{k}_3,\alpha_3|t)[f^{*}(\mathbf{k}_5,\alpha_5;\mathbf{k}_4,\alpha_4|t)\\
&+\Psi^{*}_{\mathbf{k}_5\alpha_5}(t)\Psi^{*}_{\mathbf{k}_4\alpha_4}(t)]\bigg\}-\{\text{Idem}[(\mathbf{k}_1\alpha_1)\leftrightarrow(\mathbf{k}_2\alpha_2)]\}^{*};
\end{aligned}\tag{11}$$

$$\begin{aligned}
\bigg[i\hbar\frac{d}{dt}-\hbar(\Omega_{\mathbf{k}_1\alpha_1}+\Omega_{\mathbf{k}_2\alpha_2})\bigg]f(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t)={}&\frac{1}{2V}\sum_{\mathbf{k}_3,\mathbf{k}_4}\sum_{\alpha_3,\alpha_4=1,2}\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4}[\nu_{\mathbf{k}_1-\mathbf{k}_4}(\mathcal{P}_{\mathbf{k}_1\mathbf{k}_3})_{\alpha_1\alpha_3}(\mathcal{P}_{\mathbf{k}_2\mathbf{k}_4})_{\alpha_2\alpha_4}\\
&+\nu_{\mathbf{k}_1-\mathbf{k}_3}(\mathcal{P}_{\mathbf{k}_1\mathbf{k}_4})_{\alpha_1\alpha_4}(\mathcal{P}_{\mathbf{k}_2\mathbf{k}_3})_{\alpha_2\alpha_3}][f(\mathbf{k}_3,\alpha_3;\mathbf{k}_4,\alpha_4|t)+\Psi_{\mathbf{k}_3\alpha_3}(t)\Psi_{\mathbf{k}_4\alpha_4}(t)]\\
&+\frac{1}{V}\sum_{\mathbf{k}_3,\mathbf{k}_4,\mathbf{k}_5}\sum_{\alpha_3,\alpha_4,\alpha_5=1,2}\delta_{\mathbf{k}_1+\mathbf{k}_3,\mathbf{k}_4+\mathbf{k}_5}[\nu_{\mathbf{k}_1-\mathbf{k}_5}(\mathcal{P}_{\mathbf{k}_1\mathbf{k}_4})_{\alpha_1\alpha_4}(\mathcal{P}_{\mathbf{k}_3\mathbf{k}_5})_{\alpha_3\alpha_5}\\
&+\nu_{\mathbf{k}_1-\mathbf{k}_4}(\mathcal{P}_{\mathbf{k}_1\mathbf{k}_5})_{\alpha_1\alpha_5}(\mathcal{P}_{\mathbf{k}_3\mathbf{k}_4})_{\alpha_3,\alpha_4}]\bigg\{f(\mathbf{k}_2,\alpha_2;\mathbf{k}_5,\alpha_5|t)[n(\mathbf{k}_3,\alpha_3;\mathbf{k}_4,\alpha_4|t)\\
&+\Psi^{*}_{\mathbf{k}_3\alpha_3}(t)\Psi_{\mathbf{k}_4\alpha_4}(t)]+\frac{1}{2}n^{*}(\mathbf{k}_2,\alpha_2;\mathbf{k}_3,\alpha_3|t)[f(\mathbf{k}_5,\alpha_5;\mathbf{k}_4,\alpha_4|t)\\
&+\Psi_{\mathbf{k}_5\alpha_5}(t)\Psi_{\mathbf{k}_4\alpha_4}(t)]\bigg\}+\text{Idem}[(\mathbf{k}_1\alpha_1)\leftrightarrow(\mathbf{k}_2\alpha_2)].
\end{aligned}\tag{12}$$

Here

$$n(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t)=n^*(\mathbf{k}_2,\alpha_2;\mathbf{k}_1,\alpha_1|t)$$
$$=\langle\Phi^\dagger_{\mathbf{k}_1\alpha_1}\Phi_{\mathbf{k}_2\alpha_2}\rangle_t-\Psi^*_{\mathbf{k}_1\alpha_1}(t)\Psi_{\mathbf{k}_2\alpha_2}(t), \tag{13}$$

$$f(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t)=f(\mathbf{k}_2,\alpha_2;\mathbf{k}_1,\alpha_1|t)$$
$$=\langle\Phi_{\mathbf{k}_1\alpha_1}\Phi_{\mathbf{k}_2\alpha_2}\rangle_t-\Psi_{\mathbf{k}_1\alpha_1}(t)\Psi_{\mathbf{k}_2\alpha_2}(t).$$

Equations (10)–(12) can be reduced to a simpler form in the physical situation when an external classical source is switched on and the system is prepared so that at $t=t_0$ only one macroscopically occupied coherent mode $\mathbf{k}=\mathbf{k}_0$ on the lower polariton branch is excited:

$$\Psi_{\mathbf{k}\alpha}(t_0)\propto\sqrt{V}\delta_{\alpha,1}\delta_{\mathbf{k},\mathbf{k}_0},$$
$$n(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t_0)=f(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t_0)=0. \tag{14}$$

From (10)–(12) we can find the variation of the functions $\Psi_{\mathbf{k}\alpha}(t)$ and (13) at the time moment $t=t_0+dt$:

$$d\Psi_{\mathbf{k}\alpha}\propto\delta_{\mathbf{k},\mathbf{k}_0},\quad dn(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2)=0,$$
$$df(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2)\propto\delta_{\mathbf{k}_1+\mathbf{k}_2,2\mathbf{k}_0}.$$

Therefore, the coherent polariton wave of macroscopic amplitude is a source of quantum fluctuations, described by the function $f(\mathbf{k}_1,\alpha_1;2\mathbf{k}_0-\mathbf{k}_1,\alpha_2|t)$. Substituting

$$\Psi_{\mathbf{k}\alpha}(t)=\delta_{\mathbf{k},\mathbf{k}_0}\Psi_{\mathbf{k}_0\alpha}(t),\quad n(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t)=0,$$
$$f(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t)=\delta_{\mathbf{k}_1+\mathbf{k}_2,2\mathbf{k}_0}f(\mathbf{k}_1,\alpha_1;2\mathbf{k}_0-\mathbf{k}_1,\alpha_2|t)$$

into the right-hand sides of (10)–(12) we obtain

$$d\Psi_{\mathbf{k}\alpha}\propto\delta_{\mathbf{k},\mathbf{k}_0},$$
$$dn(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t)\propto\delta_{\mathbf{k}_1,\mathbf{k}_2},df(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t)$$
$$\propto\delta_{\mathbf{k}_1+\mathbf{k}_2,2\mathbf{k}_0}.$$

This means that after the quantum fluctuations, described by abnormal distribution function $f(\mathbf{k}_1,\alpha_1;2\mathbf{k}_0-\mathbf{k}_1,\alpha_2|t)$, fluctuations that are characterized by normal (usual) distribution function $n(\mathbf{k}_1,\alpha_1;\mathbf{k}_1,\alpha_2|t)$ are excited.

Substituting

$$\Psi_{\mathbf{k}\alpha}(t)=\delta_{\mathbf{k},\mathbf{k}_0}\Psi_{\mathbf{k}_0\alpha}(t),\quad n(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t)$$
$$=\delta_{\mathbf{k}_1,\mathbf{k}_2}n(\mathbf{k}_1,\alpha_1;\mathbf{k}_1,\alpha_2|t),$$
$$f(\mathbf{k}_1,\alpha_1;\mathbf{k}_2,\alpha_2|t)=\delta_{\mathbf{k}_1+\mathbf{k}_2,2\mathbf{k}_0}f(\mathbf{k}_1,\alpha_1;2\mathbf{k}_0-\mathbf{k}_1,\alpha_2|t) \tag{15}$$

into the right-hand sides of (10)–(12) we can easily see that the left-hand sides of these equations have the same structure. Therefore, solution to the (10)–(12) under the initial conditions (14) has the form (15). This result is nothing but a consequence of the momentum conservation law in the elementary acts of polariton scattering. Further simplification of (10)–(12) can be achieved by retaining in their right-hand sides only the resonant terms corresponding to lower polariton branch. Taking into account (15) and omitting fast-oscillating terms in (10)–(12) we obtain

$$i\hbar\frac{d}{dt}\Psi_{\mathbf{k}_0}(t)=[\hbar\Omega_{\mathbf{k}_0}+\tilde{\mathcal{F}}_{\mathbf{k}_0}(t)]\Psi_{\mathbf{k}_0}(t)$$
$$+\left[\nu_{\mathrm{pol}}(\mathbf{k}_0,\mathbf{k}_0)\frac{\Psi^2_{\mathbf{k}_0}(t)}{V}+\mathcal{F}_{\mathbf{k}_0}(t)\right]\Psi^*_{\mathbf{k}_0}(t), \tag{16}$$

$$i\hbar\frac{d}{dt}n_{\mathbf{k}}(t)=f^*_{\mathbf{k}}(t)\left[\nu_{\mathrm{pol}}(\mathbf{k},\mathbf{k}_0)\frac{\Psi^2_{\mathbf{k}_0}(t)}{V}+\mathcal{F}_{\mathbf{k}}(t)\right]-f_{\mathbf{k}}(t)$$
$$\times\left[\nu_{\mathrm{pol}}(\mathbf{k},\mathbf{k}_0)\frac{\Psi^{*2}_{\mathbf{k}_0}(t)}{V}+\mathcal{F}^*_{\mathbf{k}}(t)\right], \tag{17}$$

$$i\hbar\frac{d}{dt}f_{\mathbf{k}}(t)=\left\{\left[\hbar\Omega_{\mathbf{k}}+\tilde{\nu}_{\mathrm{pol}}(\mathbf{k},\mathbf{k}_0)\frac{|\Psi_{\mathbf{k}_0}(t)|^2}{V}+\tilde{\mathcal{F}}_{\mathbf{k}}(t)\right]\right.$$
$$+\left[\hbar\Omega_{2\mathbf{k}_0-\mathbf{k}}+\tilde{\nu}_{\mathrm{pol}}(2\mathbf{k}_0-\mathbf{k},\mathbf{k}_0)\frac{|\Psi_{\mathbf{k}_0}(t)|^2}{V}\right.$$
$$\left.\left.+\tilde{\mathcal{F}}_{2\mathbf{k}_0-\mathbf{k}}(t)\right]\right\}f_{\mathbf{k}}(t)+[1+n_{\mathbf{k}}(t)+n_{2\mathbf{k}_0-\mathbf{k}}(t)]$$
$$\times\left[\nu_{\mathrm{pol}}(\mathbf{k},\mathbf{k}_0)\frac{\Psi^2_{\mathbf{k}_0}(t)}{V}+\mathcal{F}_{\mathbf{k}}(t)\right]. \tag{18}$$

Here

$$\mathcal{F}_{\mathbf{k}}(t)=\frac{1}{V}\sum_{\mathbf{k}_1}\nu_{\mathrm{pol}}(\mathbf{k},\mathbf{k}_1)f_{\mathbf{k}_1}(t)=\mathcal{F}_{2\mathbf{k}_0-\mathbf{k}}(t),$$

$$\tilde{\mathcal{F}}_{\mathbf{k}}(t)=\frac{1}{V}\sum_{\mathbf{k}_1}\tilde{\nu}_{\mathrm{pol}}(\mathbf{k},\mathbf{k}_1)n_{\mathbf{k}_1}(t)=\tilde{\mathcal{F}}^*_{\mathbf{k}}(t),$$

and

$$\nu_{\mathrm{pol}}(\mathbf{k},\mathbf{k}_1)=\frac{\nu_{\mathbf{k}-\mathbf{k}_1}}{\sqrt{(1+L^2_{\mathbf{k}})(1+L^2_{2\mathbf{k}_0-\mathbf{k}})(1+L^2_{\mathbf{k}_1})(1+L^2_{2\mathbf{k}_0-\mathbf{k}_1})}}, \tag{19}$$

$$\tilde{\nu}_{\mathrm{pol}}(\mathbf{k},\mathbf{k}_1)=\frac{\nu_0+\nu_{\mathbf{k}-\mathbf{k}_1}}{(1+L^2_{\mathbf{k}})(1+L^2_{\mathbf{k}_1})}$$

are the polariton-polariton interaction constants. According to (14), the following initial conditions should be imposed on the solution to (16)–(18):

$$n_{\mathbf{k}}(t_0)=f_{\mathbf{k}}(t_0)=0,\quad\Psi_{\mathbf{k}_0}(t_0)\neq0. \tag{20}$$

Further we shall assume $t_0=0$.

In the equilibrium problems $\mathcal{F}_{\mathbf{k}}$ and $\tilde{\mathcal{F}}_{\mathbf{k}}$ are called the order parameters and are determined from integral equations.[12,27] In the nonequilibrium problem being studied the order parameters depend on time. Therefore, their calculation involves solution of the set of nonlinear integrodifferential equations (16)–(18).

Equations (16)–(18) are invariant with respect to time inversion and do not change under the transformation $t \rightarrow -t$, $\Psi_{\mathbf{k}_0} \rightarrow \Psi^*_{\mathbf{k}_0}$, $f_{\mathbf{k}} \rightarrow f^*_{\mathbf{k}}$.

Besides additive integrals of motion displaying the conservation laws of the average values of the number of particles, energy and momentum of a closed system, Eqs. (16)–(18) possess additional integrals of motion $n_{\mathbf{k}}(t) - n_{2\mathbf{k}_0-\mathbf{k}}(t) = \text{const}$ and $|f_{\mathbf{k}}(t)|^2 - n_{\mathbf{k}}(t)[1+n_{\mathbf{k}}(t)] = \text{const}$. Using the initial conditions (20) we obtain

$$n_{\mathbf{k}}(t) = n_{2\mathbf{k}_0-\mathbf{k}}(t) = \frac{1}{2}\left[\sqrt{1+4|f_{\mathbf{k}}(t)|^2} - 1\right]. \tag{21}$$

Expressions (21) enable to exclude Eq. (17) from the set of equations (16)–(18). Note that representation of type (21) were obtained in Refs. 26 and 2.

It is easy to see that the transformation

$$f_{\mathbf{k}}(t) = \frac{\Psi^2_{\mathbf{k}_0}(t)}{|\Psi_{\mathbf{k}_0}(t)|^2}\, g_{\mathbf{k}}(t)$$

splits out of the set the equation for the phase of the condensate wave function. As a result, the set of evolution equations takes the form

$$\frac{d}{dt}\mathcal{N}_0(t) = \frac{2}{\hbar}\,\mathrm{Im}\mathcal{G}_{\mathbf{k}_0}(t)\mathcal{N}_0(t), \tag{22}$$

$$\begin{aligned} i\hbar\frac{d}{dt}g_{\mathbf{k}}(t) = \Big\{ & \hbar(\Omega_{\mathbf{k}} + \Omega_{2\mathbf{k}_0-\mathbf{k}} - 2\Omega_{\mathbf{k}_0}) + [\tilde{\mathcal{F}}_{\mathbf{k}}(t) + \tilde{\mathcal{F}}_{2\mathbf{k}_0-\mathbf{k}}(t) - 2\tilde{\mathcal{F}}_{\mathbf{k}_0}(t)] + [\tilde{\nu}_{\mathrm{pol}}(\mathbf{k},\mathbf{k}_0) + \tilde{\nu}_{\mathrm{pol}}(2\mathbf{k}_0-\mathbf{k},\mathbf{k}_0) \\ & - 2\nu_{\mathrm{pol}}(\mathbf{k}_0,\mathbf{k}_0)]\frac{|\Psi_{\mathbf{k}_0}(0)|^2}{V}\mathcal{N}_0(t) - 2\,\mathrm{Re}\mathcal{G}_{\mathbf{k}_0}(t)\Big\} g_{\mathbf{k}}(t) + \sqrt{1+4|g_{\mathbf{k}}(t)|^2}\left[\nu_{\mathrm{pol}}(\mathbf{k},\mathbf{k}_0)\frac{|\Psi_{\mathbf{k}_0}(0)|^2}{V}\mathcal{N}_0(t) + \mathcal{G}_{\mathbf{k}}(t)\right], \end{aligned} \tag{23}$$

where

$$\mathcal{G}_{\mathbf{k}}(t) = \frac{1}{V}\sum_{\mathbf{k}_1} \nu_{\mathrm{pol}}(\mathbf{k},\mathbf{k}_1) g_{\mathbf{k}_1}(t) = \mathcal{G}_{2\mathbf{k}_0-\mathbf{k}}(t),$$

and

$$\mathcal{N}_0(t) = |\Psi_{\mathbf{k}_0}(t)|^2 / |\Psi_{\mathbf{k}_0}(0)|^2$$

is the relative density of polaritons in the condensate.

IV. NUMERICAL RESULTS

The set of equations (22), (23) still remains rather complicated even for numerical methods. That is why for simplicity we neglect the dispersion of the exciton-exciton interaction constant ν. In the vicinity of exciton-photon resonance the function $L_{\mathbf{k}} \approx 1$ and, consequently, $\tilde{\nu}_{\mathrm{pol}} \approx 2\nu_{\mathrm{pol}} \approx \nu/2$.

The initial conditions (20) are the same for all values of the wave vector $\mathbf{k}$. Therefore, as follows from Eqs. (22), (23), when there is no dispersion of the polariton-polariton interaction constants, g depends on $\mathbf{k}$ via the functions $\Omega_{\mathbf{k}} + \Omega_{2\mathbf{k}_0-\mathbf{k}} - 2\Omega_{\mathbf{k}_0}$:

$$g_{\mathbf{k}} \equiv g(\Omega_{\mathbf{k}} + \Omega_{2\mathbf{k}_0-\mathbf{k}} - 2\Omega_{\mathbf{k}_0}, t).$$

Using the identical transformation

$$\begin{aligned} \frac{1}{V}\sum_{\mathbf{k}} g(\Omega_{\mathbf{k}} + \Omega_{2\mathbf{k}_0-\mathbf{k}} - 2\Omega_{\mathbf{k}_0}, t) &= \frac{1}{V}\sum_{\mathbf{k}} \int dx\, g(\Omega_{\mathbf{k}} + \Omega_{2\mathbf{k}_0-\mathbf{k}} - 2\Omega_{\mathbf{k}_0}, t)\,\delta(\Omega_{\mathbf{k}} + \Omega_{2\mathbf{k}_0-\mathbf{k}} - 2\Omega_{\mathbf{k}_0} - x) \\ &= \int dx\, g(x,t)\frac{1}{V}\sum_{\mathbf{k}} \delta(\Omega_{\mathbf{k}} + \Omega_{2\mathbf{k}_0-\mathbf{k}} - 2\Omega_{\mathbf{k}_0} - x), \end{aligned}$$

and moving to the dimensionless variables

$$T=\lambda t, w_{\mathbf{k}}=\lambda^{-1}(\Omega_{\mathbf{k}}+\Omega_{2\mathbf{k}_0-\mathbf{k}}-2\Omega_{\mathbf{k}_0}),$$

$$\lambda=\frac{\nu}{2\hbar}\frac{|\Psi_{\mathbf{k}_0}(0)|^2}{V}, \tag{24}$$

we obtain the set of equations

$$\frac{d}{dT}\mathcal{N}_0(T)=2\ \mathrm{Im}G(T)\mathcal{N}_0(T), \tag{25}$$

$$i\frac{\partial}{\partial T}g(w,T)=[w+\mathcal{N}_0(T)-2\ \mathrm{Re}G(T)]g(w,T)+\sqrt{1+4|g(w,T)|^2}\left[\frac{1}{2}\mathcal{N}_0(T)+G(T)\right], \tag{26}$$

where

$$G(T)=\int dw\rho(w)g(w,T),$$

$$\rho(w)=\frac{1}{2|\Psi_{\mathbf{k}_0}(0)|^2}\sum_{\mathbf{k}}\delta(w_{\mathbf{k}}-w). \tag{27}$$

Further the parameters of polaritons formed by mixing of photons and 1A excitons in CdS single crystal are used for numerical estimation: $\epsilon_B=9.3$, $\hbar\omega^{\perp}=2.55$ eV, $\eta/\hbar=1.1\times10^{14}c^{-1}$, $m_\perp=0.89m_0$ (m_0 is the free electron mass), and $m_\perp=2.85m_0$.[33] The model of isotropic parabolic exciton band with effective mass $m=(m_\perp^2m_\parallel)^{1/3}$. The effective Bohr radius of the 1A exciton in CdS is $a_{\mathrm{ex}}=28$ Å.[34] The exciton ionization energy is $I_{\mathrm{ex}}=27$ meV.[35] The value of the exciton-exciton interaction constant is evaluated by the formula $\nu\equiv\nu_0=(26\pi/3)I_{\mathrm{ex}}a_{\mathrm{ex}}^3$ (Ref. 36) and makes 4.3×10^{-33} erg cm^3.

At the initial stage of evolution of the system when a considerable portion of polaritons is still in the condensate and the number of noncondensate polaritons is rather small we can omit the terms containing $G(T)$ in Eqs. (25), (26). The solution to the obtained set of equations has the form

$$\mathcal{N}_0(T)=\mathrm{const}=1$$

(the influence of a small portion of noncondensate polaritons on the condensate is not taken into account), and

$$n(w,T)=\begin{cases}-\dfrac{1}{w(w+2)}\sinh^2\left[\dfrac{T}{2}\sqrt{-w(w+2)}\right] & \text{for } -2\leqslant w\leqslant 0\\[2ex] \dfrac{1}{w(w+2)}\sin^2\left[\dfrac{T}{2}\sqrt{w(w+2)}\right] & \text{for } w<-2, w>0\,.\end{cases} \tag{28}$$

Thus, at the initial stage of evolution the excitation of noncondensate polaritons takes place in the range $-2\leqslant w\leqslant 0$ where the distribution function $n(w)$ has the form of a symmetrical bell with a maximum at $w=-1$. While moving away from the above-mentioned region the distribution function decreases with oscillations.

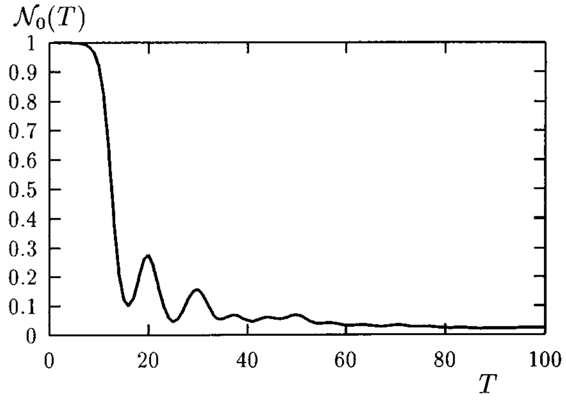

FIG. 1. Dependence of a relative polariton density in the condensate on dimensionless time T.

The numerical solution of the equation set (25), (26) was made in two stages. First, the explicit form of the function $\rho(w)$ for different values of condensate density $|\Psi_{\mathbf{k}_0}(0)|^2/V$ and its wave vector $\mathbf{k}_0$ magnitude was found. Further, the function obtained was used to integrate the evolution equations (25), (26). The result of the integration for $|\mathbf{k}_0|=3.6\times10^5$ cm^{-1} are shown in Figs. 1–3. This choice of $|\mathbf{k}_0|$ corresponds to the exciton-photon resonance region located a bit below the exciton band bottom.

The decay of the nonequilibrium polariton condensate is shown in Fig. 1. Before the moment of dimensionless time $T\approx7$ the condensate depletion proceeds rather slowly. Then a drastic fall takes place. As a result, by the moment $T\approx15$ only 10% of the initial polariton number remain in the condensate. The further condensate decay is accompanied by oscillations. This leads to the partial restoration of the condensate (up to 30% at $T\approx20$). The oscillations do not occur if in Eq. (26) the function $G(T)$, describing the integral influence of all pairs of noncondensate polaritons with the same total momentum on each individual pair, is neglected. Thus, the occurrence of oscillations is due to correlation of the states of individual pairs of noncondensate polaritons. By the moment $T\approx50$ the oscillations disappear and further con-

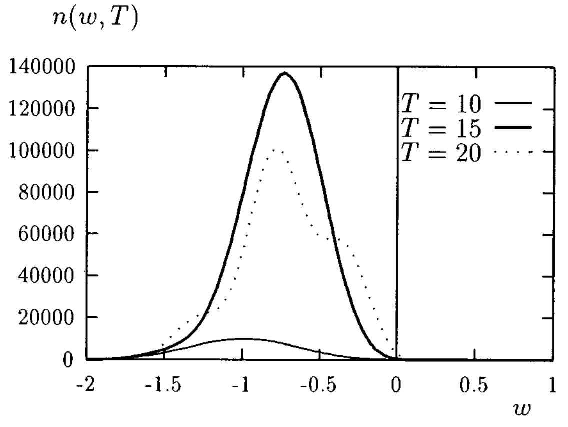

FIG. 2. The distribution function of noncondensate polaritons at T=10,15,20.

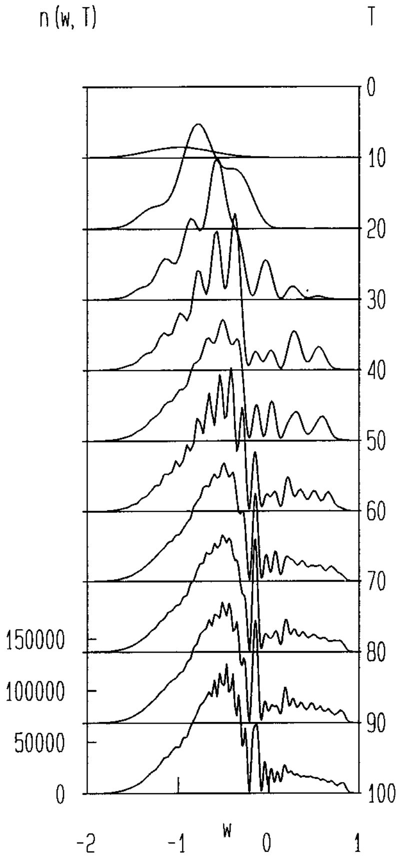

FIG. 3. The distribution function of noncondensate polaritons at different moments of dimensionless time T. The function scale is shown by the marks in the left vertical axis.

densate evolution has the form of slow monotonous decay. These results cannot be obtained by the introduction of phenomenological constants in the dynamic equations.

Figure 2 depicts the distribution function of noncondensate polaritons at time moments in the interval where the polariton condensate undergoes the most rapid and significant changes. The curve corresponding to the moment of time $T=10$ as well as the curve described by the function (28) has the form of a bell with the maximum at $w=-1$. By the moment $T=15$ the distribution function without changing its symmetrical form increases significantly its value and its maximum shifts to greater energies of noncondensate polaritons. An increase of the number of noncondensate polaritons by more than an order of magnitude corresponds to the presence of the local minimum of $\mathcal{N}_0(T)$ function at $T\approx 15$. When $T=20$ the distortion of the distribution function takes place and the area under the curve diminishes. This is in accordance with the local maximum of the $\mathcal{N}_0(T)$ at $T\approx 20$. Thus, the distribution function is ''breathing'' in conformity with the variation of condensate density.

Further evolution of the distribution function is shown in Fig. 3. At the moment of time $T=30$ its contour gets the oscillating character. The oscillations arise not only in the dependence of the distribution function on parameter w but in its dependence on dimensionless time T (unfortunately we could not demonstrate this in the picture). This means that if, e.g., the distribution function at the moment $T\approx 40$ possesses a local maximum at $w=-1$ it can possess at this point a local minimum at another moment of time. Later on, the frequency oscillations become denser, smaller, and transform into chaotic ripples on the smooth contour of the distribution function.

From the moment $T\approx 30$ a short-wave wing begins to arise in the distribution function curve. By the moment of time $T=100$ the function localizes in the interval $-2<w<1$. Very important is the enhancement and then the partial extinction of the sharp extra peak localized within the region $-0.22<w<-0.08$ with a maximum at $w=-0.15$. The time interval where the extra peak arises and exists is characterized by small values of condensate density and its relatively slow change (see Figs. 1 and 3). The extra peak does not change its position with time and reaches its maximum at $T\approx 80$.

Under the change of the initial condensate density $|\Psi_{\mathbf{k}_0}(0)|^2/V$ only the scale of shown dependences changes due to the corresponding transformation of dimensionless arguments (24). This result follows from the very weak dependence of the solution of Eqs. (25), (26) on the initial condensate density via the function $\rho(w)$ involved in the definition of the $G(T)$ function. According to Figs. 2 and 3, $g(w,T)$ differs from zero only at $-2<w<1$. The numerical results show that in that region $\rho(w)$ does not practically depend on its argument due to peculiarities of low polariton dispersion in the vicinity of exciton-photon resonance. This enables us to replace the function $\rho(w)$ by $\rho(0)$ in the definition (27). As a result we obtain $G(T)\approx\rho(0)\int dwg(w,T)$. But according to (27) and (24) $\rho(0)$ does not depend on the initial condensate density and is determined only by the value of wave vector $\mathbf{k}_0$ under the given crystal parameters. The value $\rho(0)\sim 6.8\times 10^{-6}$ corresponds to the value $|\mathbf{k}_0|=3.6\times 10^5\ \mathrm{cm}^{-1}$.

V. CONCLUSIONS

Therefore, the significant condensate depletion takes place already at the before-kinetic stage of the system evolution. Within a short time $t_*\sim 15\lambda^{-1}$ the condensate density diminishes 10 times compared to the initial density. For CdS single crystals and initial condensate density 10^{16}–$10^{18}\ \mathrm{cm}^{-3}$ the time t_* equals to 120–1.2 ps. The short time of the polariton condensate decay makes problematic the possibility of observing such coherent nonlinear phenomena as, e.g., the phenomenon of self-induced transparency and creation of solitons in exciton spectral range.[37–40]

Note the irreversible-in-time character of the described solution of the time-reversible equations.[41]

We would like to point out also that in Ref. 42 the steady-state solution $|\Psi_{\mathbf{k}_0}(t)|^2=\mathrm{const}$, $n_{\mathbf{k}}(t)\propto\delta(E_{\mathbf{k}}+E_{2\mathbf{k}_0-\mathbf{k}}-2E_{\mathbf{k}_0})$, $f_{\mathbf{k}}(t)\propto\delta(E_{\mathbf{k}}+E_{2\mathbf{k}_0-\mathbf{k}}-2E_{\mathbf{k}_0})$ to Eqs. (16)–(18) has been obtained ($E_{\mathbf{k}}$ is the renormalized energy of the polariton). Substitution of this solution in the expressions for the total average energy and total average number of particles shows that this solution cannot be obtained under the initial conditions (20). But the state of the polariton system corresponding to this solution can be arranged by means of steady laser action on a semiconductor.[43]

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[1] V. M. Galitskii and V. F. Elesin, *Resonant Interaction of the Electromagnetic Fields with Semiconductors* (Energoatomizdat, Moscow, 1986).

[2] S. Schmitt-Rink, D. S. Chemla and H. Haug, Phys. Rev. B **37**, 941 (1988).

[3] L. V. Keldysh, Phys. Status Solidi B **188**, 11 (1995).

[4] A. L. Ivanov and L. V. Keldysh, Zh. Éksp. Teor. Fiz. **84**, 404 (1983) [Sov. Phys. JETP **57**, 234 (1983)].

[5] M. V. Lebedev, Zh. Éksp. Teor. Fiz. **101**, 957 (1992).

[6] V. N. Chumash, I. A. Cojocaru, I. I. Dobynde, V. N. Pitei, and M. I. Shmigliuk, in *Proceedings of the International Conference on Excitons 84* (Güstrow, GDR, 1984), pp. 174–177.

[7] M. I. Shmigliuk and V. N. Pitei, *Coherent Polaritons in Semiconductors* (Ştiinţa, Kishinev, 1989).

[8] N. N. Bogoliubov, Vestnik MGU **7**, 43 (1947); see also N. N. Bogoliubov, *Selected Works* (Naukova Dumka, Kiev, 1970), Vol. 2, pp. 242–257.

[9] L. D. Landau and E. M. Lifshitz, *Course of Theoretical Physics* (Pergamon, New York, 1965), Vol. 3.

[10] V. R. Misko, S. A. Moskalenko, and M. I. Shmigliuk, Fiz. Tverd. Tela **35**, 3213 (1993) [Sov. Phys. Solid State **43**, 287 (1994)].

[11] A. I. Bobrysheva, S. A. Moskalenko, M. I. Shmigliuk, and S. S. Russu, Phys. Status Solidi B **190**, 481 (1995).

[12] J. R. Schrieffer, *Theory of Superconductivity* (Addison-Wesley, Menlo Park, CA, 1964).

[13] N. N. Bogoliubov (unpublished); see also N. N. Bogoliubov, *Selected Works* (Naukova Dumka, Kiev, 1971), Vol. 3, pp. 174–243.

[14] N. N. Bogoliubov, Jr. and B. I. Sadovnikov, *Some Questions of Statistical Mechanics* (Vysshaya Shkola, Moscow, 1975).

[15] V. N. Popov and V. S. Yarunin, *Collective Effects in Quantum Statistics of Radiation and Matter* (Izd. LGU, Leningrad, 1985).

[16] S. A. Moskalenko, A. H. Rotaru, and Yu. M. Shvera, in *Laser Optics of Condensed Matter*, edited by J. L. Birman, H. Z. Cummins, and A. A. Kaplyanskii (Plenum, New York, 1988), pp. 331–336.

[17] V. R. Misko, S. A. Moskalenko, A. H. Rotaru, and Yu. M. Shvera, Phys. Status Solidi B **159**, 477 (1990).

[18] I. V. Beloussov and Yu. M. Shvera, Phys. Status Solidi B **139**, 91 (1990).

[19] I. V. Beloussov and Yu. M. Shvera, Z. Phys. B **90**, 51 (1993).

[20] L. V. Keldysh, Zh. Éksp. Teor. Fiz. **47**, 1515 (1964) [Sov. Phys. JETP **20**, 1018 (1965)].

[21] L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (Benjamin, New York, 1962).

[22] I. V. Beloussov and Yu. M. Shvera, Theor. Math. Phys. **85**, 1177 (1991).

[23] D. N. Zubarev, *Nonequilibrium Statistical Thermodynamics* (Plenum, New York, 1974).

[24] L. V. Keldysh, in *Problems of Theoretical Physics, A Memorial Volume to Igor E. Tamm*, edited by V. I. Ritus (Nauka, Moscow, 1972), pp. 433–444.

[25] A. I. Akhiezer and S. V. Peletminskii, *Methods of Statistical Physics* (Nauka, Moscow, 1977).

[26] S. Schmitt-Rink and D. S. Chemla, Phys. Rev. Lett. **57**, 2752 (1986).

[27] C. Comte and G. Mahler, Phys. Rev. B **38**, 10 517 (1988).

[28] A. I. Bobrysheva and S. A. Moskalenko, Fiz. Tverd. Tela, **25**, 3282 (1983) [Sov. Phys. Solid State **25**, 1891 (1983)].

[29] A. I. Bobrysheva, S. A. Moskalenko, and Yu. M. Shvera, Phys. Status Solidi B **147**, 711 (1988).

[30] L. V. Keldysh, Solid State Commun. **84**, 37 (1992).

[31] L. V. Keldysh, Phys. Status Solidi B **173**, 119 (1992).

[32] A. I. Bobrysheva, S. A. Moskalenko, and Xoang Ngok Cam, Zh. Éksp. Teor. Fiz. **103**, 301 (1993) [Sov. Phys. JETP **76**, 163 (1993)].

[33] E. S. Koteles, in *Excitons*, edited by E. I. Rashba and M. D. Sturge (North-Holland, Amsterdam, 1982).

[34] B. S. Wang and J. L. Birman, Phys. Rev. B **42**, 9609 (1990).

[35] W. F. Brinkman, T. M. Rice, and B. Bell, Phys. Rev. B **8**, 1570 (1966).

[36] A. I. Bobrysheva, M. F. Miglei, and M. I. Shmigliuk, Phys. Status Solidi B **53**, 71 (1972).

[37] J. Goll and H. Haken, Opt. Commun. **24**, 1 (1978).

[38] J. Goll and H. Haken, Phys. Rev. A **18**, 2241 (1978).

[39] S. N. Belkin, P. I. Khadzhi, S. A. Moskalenko, and A. H. Rotaru, J. Phys. C **14**, 4109 (1981).

[40] P. I. Khadzhi, S. A. Moskalenko, A. H. Rotaru, and G. D. Shibarshina, Phys. Status Solidi B **114**, K25 (1982).

[41] G. M. Zaslavskii, *Stochasticity of Dynamic Systems* (Nauka, Moscow, 1984) (in Russian).

[42] I. V. Beloussov and Yu. M. Shvera, in *Bose-Einstein Condensation*, edited by A. Griffin, D. W. Snoke, and S. Stringari (Cambridge University Press, New York, 1995), pp. 513–518.

[43] V. L. Safonov, Physica A **188**, 675 (1992).