

Nonlinear intrinsic-nutation of high-density polaritons in direct-gap semiconductors

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(Submitted 28 July 1995; resubmitted 16 November 1995)
Zh. Éksp. Teor. Fiz. **109**, 1806–1822 (May 1996)

We use the microscopic approach to study the evolution of a nonequilibrium polariton condensate and show that as a result of polariton–polariton scattering the condensate becomes considerably depleted in a very short time interval whose value is determined by the initial density of the condensate. Disintegration of the condensate has the form of nonlinear damped oscillations. We study the dynamics of the distribution function of extracondensate polaritons and show that in the initial stage of the system's evolution the distribution function has the shape of a symmetric bell. With the passage of time oscillations appear in the envelope of the distribution function, and these are gradually transformed into shallow chaotic ripples. Formation of the short-wave wing of the distribution function is also observed. Attention is focused on the ignition, followed by partial extinction, of an additional sharp peak, which appears in a time interval characterized by low values of the density of the polariton condensate and slow variations of this density. © 1996 American Institute of Physics. [S1063-7761(96)02105-1]

1. INTRODUCTION

The classical approach is used in a large number of studies devoted to the spatial–temporal evolution of intense laser radiation in resonant media. This approach presupposes that the photon and material fields are macrofilled coherent modes, or condensates, interacting with each other. It is assumed that, first, disintegration of the condensates takes place exponentially and, second, that the lifetime of a condensate far exceeds the time of the nonlinear coherent process under investigation. The classical approach was used, in particular, by Moskalenko *et al.*¹ to study the phenomena of self-induced transparency and nutation in the excitonic range of the spectrum.

We believe, however, that the microscopic approach is more consistent. In it the dynamics of the interacting condensate modes and quantum fluctuations appearing in the system as a result of real quasiparticle scattering processes is described in a unified manner. In other words, we must allow for (a) the interaction of the condensates with each other, (b) the excitation of extracondensate quasiparticles, and (c) the effect of the extracondensate particles on the condensates. In the present paper the microscopic approach is used to describe the temporal evolution of a system of high-density polaritons that initially was coherent.

As is known,² when an electromagnetic wave whose frequency is in resonance with a specified exciton energy level acts on a direct-gap semiconductor, a coherent polariton wave with a wave vector $\mathbf{k}_0 \neq 0$ is excited in the crystal, or a nonequilibrium polariton condensate is formed. As the wave propagates through the crystal and various scattering processes take place, the initial polariton wave may lose its coherence, the condensate can be depleted either completely or partially, polaritons with wave vectors $\mathbf{k} \neq \mathbf{k}_0$ may be excited, etc.

At high crystal excitation energies, polariton scattering associated with exciton–exciton collisions dominates. Inter-

est in this scattering mechanism has been stimulated by recent experimental studies³ and by a large body of intriguing physical results obtained in theoretical studies of the dynamic and kinetic changes emerging in a polariton system as a result of the interaction between excitons (see, e.g., Refs. 4 and 5).

According to Refs. 5 and 6, when exciton–exciton collisions are taken into account, the most interesting situation from the standpoint of physics is the one in which the coherent excitation of polaritons is achieved in a certain spectral range for which the laws of energy and momentum conservation allow for real processes of two-photon escape of polaritons from the condensate. These processes lead to instabilities in the fully condensed state of the polariton system. The existence of such a range near an isolated exciton resonance is related to the special features of the polariton dispersion law.

Piteř *et al.*⁶ studied the energy spectrum of extracondensate polaritons resulting from the decay of a coherent polariton wave. They found that \mathbf{k} -space has regions in which, as the researchers believe, no energy spectrum can exist.

We note at this point that the research in Ref. 6 is based on a model which, by its formal properties, is similar to that used by Bogolyubov⁷ to study the equilibrium system of a weakly nonideal Bose gas. In the nonequilibrium physical situation examined in Ref. 6, in which the polariton condensate is in the process of disintegration accompanied by excitation of a large number of extracondensate polariton, the model can be thought of as reflecting the physical essence of the problem correctly only in the initial stage of the decay of the condensed state of the system, when the number of polaritons in the condensate is still considerably greater than the total number of extracondensate polaritons. This stage, however, is essentially time-dependent, which is proved by the instability of the condensate. But studying the energy spectrum presupposes finding the stationary states of the system (see Ref. 8, p. 28). For this reason we believe that the

results of Refs. 5 and 6 based on the specified model and referring to the energy spectrum of the system cannot be considered justified.

Strictly speaking, the largely time-dependent nature of the processes taking place in the system requires using the methods of nonequilibrium statistical mechanics. Here the derivation of equations describing the disintegration of the polariton condensate and the excitation of quantum fluctuations possesses several special features related to the existence of degeneracy in the system. Since the total energy and the total momentum of two extracondensate polaritons may prove to be equal to, respectively, the energy and momentum of two polaritons in the condensate, we are forced to deal with the degeneracy of two-particle states. In addition, the mere presence of a condensate implies degeneracy caused by the condensate's macroscopic amplitude.⁹

To describe a system with degeneracy in a meaningful way we must introduce, in addition to the ordinary (normal) distribution function $N_{\mathbf{k}} = \langle \hat{\Phi}_{\mathbf{k}}^+ \hat{\Phi}_{\mathbf{k}} \rangle$, anomalous functions $\Psi_{\mathbf{k}_0} = \langle \hat{\Phi}_{\mathbf{k}_0} \rangle$ and $F_{\mathbf{k}} = \langle \hat{\Phi}_{\mathbf{k}} \hat{\Phi}_{2\mathbf{k}_0 - \mathbf{k}} \rangle$ (see Ref. 10). Here $\hat{\Phi}_{\mathbf{k}}^+$ ($\hat{\Phi}_{\mathbf{k}}$) is the Bose creation (annihilation) operator of a polariton in a state with a wave vector \mathbf{k} on the lower polariton branch. The appearance of anomalous averages means that the selection rules related to the gauge invariance of the system are violated.¹⁰⁻¹² In our problem gauge symmetry breaking appears because of the noninvariant initial condition related to the assumption that initially (at time $t = t_0$) the system contained a condensate.

An attempt to derive the kinetic equations for a system of polaritons excited in a semiconductor by an external classical source acting in a time-independent manner was made in Refs. 13 and 14. The researchers, however, did not account for the degeneracy of two-particle states in the system. In particular, no anomalous distribution function $F_{\mathbf{k}}$ was introduced. No wonder then that the equations they obtained contained a nonphysical singularity.

Kinetic equations describing the evolution of partially coherent polaritons and allowing for degeneracy were found in Refs. 15 and 16 by Keldysh's method¹⁷ formulated in terms of functionals. The equations coincide with those obtained by the same authors in Ref. 18 by the method of a nonequilibrium statistical operator¹⁹ and contain no nonphysical singularities. We would also like to mention the reports in Refs. 20 and 21, which contain the most thorough description of Refs. 15, 16, and 18.

According to Refs. 16 and 18, the kinetics of partially coherent polaritons is described in the Born approximation by a system of nonlinear integro-differential equations for the coherent part of the polariton field $\Psi_{\mathbf{k}_0}$ and the normal and anomalous distribution functions $n_{\mathbf{k}} = N_{\mathbf{k}} - \delta_{\mathbf{k}, \mathbf{k}_0} |\Psi_{\mathbf{k}_0}|^2$ and $f_{\mathbf{k}} = F_{\mathbf{k}} - \delta_{\mathbf{k}, \mathbf{k}_0} \Psi_{\mathbf{k}_0}^2$, respectively. In the absence of quantum fluctuations described by the functions $n_{\mathbf{k}}$ and $f_{\mathbf{k}}$, the equations for these function become identities, and the equations for $\Psi_{\mathbf{k}\alpha}$ (α is the number of the polariton branch) are in this case equivalent to those obtained in Ref. 22 for a system of interacting coherent excitons and photons. In another special case in which $\Psi_{\mathbf{k}_0} = 0$ and $f_{\mathbf{k}} = 0$, an ordinary kinetic equation follows from the results of Ref. 16 for the

distribution function $N_{\mathbf{k}}$ given, for instance, in Ref. 23.

The right-hand sides of the kinetic equations suggested in Refs. 16-18 contain terms linear in the exciton-exciton coupling constant $\nu > 0$ and terms proportional to ν^2 . The first correspond to the self-consistent field approximation, which means ignoring higher-order correlation functions. This approximation allows for the two-photon escape of polaritons from the condensate, reverse processes, and the transformation of a created pair of extracondensate polaritons into another pair with the same total momentum. These terms describe the fastest processes in the system and are sufficient for studying the early (pre-kinetic) stage in the evolution of the system.

The terms proportional to ν^2 allow for scattering processes in which only one polariton belongs to the condensate. For this reason they are nonzero only when the system has extracondensate polaritons. Such terms describe slower variations (compared to those described by the terms proportional to ν) of the system's characteristics and become important only in the kinetic stage of the evolution. Our study of the temporal evolution of a system of nonequilibrium polaritons is based entirely on the self-consistent field approximation.

2. THE SYSTEM HAMILTONIAN

We start with the simplest Hamiltonian^{4,24}

$$\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}}, \quad (1)$$

$$\hat{H}_0 = \sum_{\mathbf{k}} \sum_{\alpha_1, \alpha_2=1,2} (h_{\mathbf{k}})_{\alpha_1 \alpha_2} \hat{\varphi}_{\mathbf{k}\alpha_1}^+ \hat{\varphi}_{\mathbf{k}\alpha_2},$$

$$h_{\mathbf{k}} = \hbar \omega_{\mathbf{k}}^{\perp} \left(\frac{1 + \tau_3}{2} \right) + \hbar \omega_{\mathbf{k}} \left(\frac{1 - \tau_3}{2} \right) + \eta_{\mathbf{k}} \tau_2, \quad (2)$$

$$\hat{H}_{\text{int}} = \frac{1}{2V} \sum_{\mathbf{k}_1, \dots, \mathbf{k}_4} \sum_{\alpha_1, \dots, \alpha_4=1,2} \nu_{\mathbf{k}_1 - \mathbf{k}_4} \delta_{\mathbf{k}_1 + \mathbf{k}_2, \mathbf{k}_3 + \mathbf{k}_4}$$

$$\times \left(\frac{1 + \tau_3}{2} \right)_{\alpha_1, \alpha_3} \left(\frac{1 + \tau_3}{2} \right)_{\alpha_2, \alpha_4}$$

$$\times \hat{\varphi}_{\mathbf{k}_1 \alpha_1}^+ \hat{\varphi}_{\mathbf{k}_2 \alpha_2}^+ \hat{\varphi}_{\mathbf{k}_3 \alpha_3} \hat{\varphi}_{\mathbf{k}_4 \alpha_4}. \quad (3)$$

The creation (annihilation) operators $\hat{\varphi}_{\mathbf{k}\alpha}^+$ ($\hat{\varphi}_{\mathbf{k}\alpha}$) of an exciton (at $\alpha = 1$) or photon (at $\alpha = 2$) in a state with wave vector \mathbf{k} obey the Bose commutation relations:

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

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

For the Pauli matrices τ_1 , τ_2 , and τ_3 we employ the standard representation.¹⁷ The frequencies $\omega_{\mathbf{k}}$ and $\omega_{\mathbf{k}}^{\perp}$ are defined by $\omega_{\mathbf{k}} = c|\mathbf{k}| \epsilon_B^{-1/2}$ and $\omega_{\mathbf{k}}^{\perp} = \omega_{\mathbf{k}} + \hbar \mathbf{k}^2 / 2m$, where c is the speed of light in vacuum, $\hbar \omega_{\mathbf{k}}^{\perp}$ is the energy needed to form an exciton in the band $\hbar \omega_{\mathbf{k}}^{\perp}$, and m is the exciton effective mass. The background dielectric constant ϵ_B allows for the contribution to the total dielectric constant of the crystal of all excitations except for the excitons in the isolated band $\hbar \omega_{\mathbf{k}}^{\perp}$.

The Hamiltonian (1) describes a system of interacting dipole-active excitons and transverse linearly polarized photons in an infinite crystal ($V \rightarrow \infty$, where V is the quantization volume) near an isolated excitonic resonance $\omega_{\mathbf{k}} \approx \omega_{\mathbf{k}}^{\perp}$ (see Ref. 24).

In other words, we assume that the photon frequency $\hbar\omega_{\mathbf{k}}$, the exciton energy $\hbar\omega_{\mathbf{k}}^{\perp}$, and the minimum energy gap $\Delta\mathcal{E}$ separating the exciton band $\hbar\omega_{\mathbf{k}}$ from any other bands are linked by the following relationships:

$$\hbar|\omega_{\mathbf{k}} - \omega_{\mathbf{k}}^{\perp}| \ll \hbar\omega_{\mathbf{k}}^{\perp}, \Delta\mathcal{E}.$$

We also assume that the exciton-photon ($\eta_{\mathbf{k}} = \eta_{-\mathbf{k}}$) and exciton-exciton ($\nu_{\mathbf{k}} = \nu_{-\mathbf{k}}$) coupling constants are fairly small:

$$0 < \eta_{\mathbf{k}} \ll \omega_{\mathbf{k}}^{\perp}, \quad 0 < (\nu_{\mathbf{k}}/\hbar)\bar{n} \ll \omega_{\mathbf{k}}^{\perp}$$

(here \bar{n} is the mean exciton number density in the system). The first inequality makes it possible to keep only the resonance terms in the Hamiltonian (2), while the second allows considering the exciton-exciton coupling a small perturbation. If the condition for resonance, $\omega_{\mathbf{k}} \approx \omega_{\mathbf{k}}^{\perp}$, is met, the weak frequency dependence of ϵ_B can be ignored.

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

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

by introducing the Bose polariton operators $\hat{\Phi}_{\mathbf{k}\alpha}$ and $\hat{\Phi}_{\mathbf{k}\alpha}^{\dagger}$ via the transformation

$$\hat{\Phi}_{\mathbf{k}\alpha_1} = \sum_{\alpha_2=1,2} (U_{\mathbf{k}})_{\alpha_1\alpha_2} \hat{\Phi}_{\mathbf{k}\alpha_2}, \quad U_{\mathbf{k}} = \frac{1 - i\tau_1\Lambda_{\mathbf{k}}}{\sqrt{1 + \Lambda_{\mathbf{k}}^2}}.$$

Here the function $\Lambda_{\mathbf{k}}$ can be found by solving the equation $\eta_{\mathbf{k}}\Lambda_{\mathbf{k}}^2 + \hbar(\omega_{\mathbf{k}} - \omega_{\mathbf{k}}^{\perp})\Lambda_{\mathbf{k}} - \eta_{\mathbf{k}} = 0$. In the polariton representation,

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

where

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

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

3. THE SELF-CONSISTENT FIELD APPROXIMATION

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

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

and the average value of an arbitrary dynamical quantity is defined as $\langle A \rangle_t = \text{Tr} \check{\rho} \check{A}(t)$. Here $\check{\rho}$ is the density matrix defining the distribution of polaritons initially (at $t=t_0$), and the ‘‘check’’ sign on operators denotes the Heisenberg picture.

When there is a condensate in the system, the average value of the polariton annihilation operator

$$\Psi_{\mathbf{k}\alpha}(t) = \langle \check{\Phi}_{\mathbf{k}\alpha}(t) \rangle_t \quad (7)$$

is nonzero. Because of the interaction between polaritons this average value varies with time. Out of the operator $\check{\Phi}_{\mathbf{k}\alpha}(t)$ we take the coherent part:

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

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

We substitute (8) into the system Hamiltonian $\check{H} = \check{H}_0 + \check{H}_{\text{int}}$ and represent the Hamiltonian as $\check{H} = \check{H}_1(t) + \check{H}_2(t)$, where the operator $\check{H}_1(t)$ contains only terms that are linear or bilinear in $\check{\chi}_{\mathbf{k}\alpha}(t)$ and $\check{\chi}_{\mathbf{k}\alpha}^{\dagger}(t)$. The operator $\check{H}_2(t)$ contains products of three or four operators $\check{\chi}_{\mathbf{k}\alpha}(t)$ and $\check{\chi}_{\mathbf{k}\alpha}^{\dagger}(t)$.

We now allow for $\check{H}_2(t)$ in the self-consistent field approximation. To this end we make the formal substitution:

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

As a result, $\check{H}_2(t)$ and hence \check{H} acquire the same operator structure as $\check{H}_1(t)$. If we now go from the operators $\check{\chi}_{\mathbf{k}\alpha}(t)$ to the original operators $\check{\Phi}_{\mathbf{k}\alpha}(t)$ in \check{H} , we obtain

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

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

Combining the expression for the Hamiltonian (9) and the equations of motion (6), we find the system of equations describing the temporal evolution of a system of partially coherent polaritons in the self-consistent field approximation. If we introduce the distribution functions

$$n(\mathbf{k}_1, \alpha_1; \mathbf{k}_2, \alpha_2|t) = \langle \Phi_{\mathbf{k}_1 \alpha_1}^+ \Phi_{\mathbf{k}_2 \alpha_2} \rangle_t - \Psi_{\mathbf{k}_1 \alpha_1}^*(t) \Psi_{\mathbf{k}_2 \alpha_2}(t), \quad (10)$$

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

we get

$$\begin{aligned} & \left[i\hbar \frac{d}{dt} - \hbar \Omega_{\mathbf{k}_1 \alpha_1} \right] \Psi_{\mathbf{k}_1 \alpha_1}(t) \\ &= \frac{1}{V} \sum_{\mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4} \sum_{\alpha_2, \alpha_3, \alpha_4=1,2} \delta_{\mathbf{k}_1 + \mathbf{k}_2, \mathbf{k}_3 + \mathbf{k}_4} \\ & \times [\nu_{\mathbf{k}_1 - \mathbf{k}_4}(\mathcal{P}_{\mathbf{k}_1 \mathbf{k}_3})_{\alpha_1 \alpha_3}(\mathcal{P}_{\mathbf{k}_2 \mathbf{k}_4})_{\alpha_2 \alpha_4} \\ & + \nu_{\mathbf{k}_1 - \mathbf{k}_3}(\mathcal{P}_{\mathbf{k}_1 \mathbf{k}_4})_{\alpha_1 \alpha_4}(\mathcal{P}_{\mathbf{k}_2 \mathbf{k}_3})_{\alpha_2 \alpha_3}] \\ & \times \left[\frac{1}{2} \Psi_{\mathbf{k}_2 \alpha_2}^*(t) \Psi_{\mathbf{k}_3 \alpha_3}(t) \Psi_{\mathbf{k}_4 \alpha_4}(t) \right. \\ & + n(\mathbf{k}_2, \alpha_2; \mathbf{k}_3, \alpha_3|t) \Psi_{\mathbf{k}_4 \alpha_4}(t) \\ & \left. + \frac{1}{2} \Psi_{\mathbf{k}_2 \alpha_2}^*(t) f(\mathbf{k}_3, \alpha_3; \mathbf{k}_4, \alpha_4|t) \right], \quad (11) \end{aligned}$$

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

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

$$\begin{aligned} & + \Psi_{\mathbf{k}_3 \alpha_3}(t) \Psi_{\mathbf{k}_4 \alpha_4}(t)] \\ & + \frac{1}{V} \sum_{\mathbf{k}_3, \mathbf{k}_4, \mathbf{k}_5} \sum_{\alpha_3, \alpha_4, \alpha_5=1,2} \delta_{\mathbf{k}_1 + \mathbf{k}_3, \mathbf{k}_4 + \mathbf{k}_5} \\ & \times [\nu_{\mathbf{k}_1 - \mathbf{k}_5}(\mathcal{P}_{\mathbf{k}_1 \mathbf{k}_4})_{\alpha_1 \alpha_4}(\mathcal{P}_{\mathbf{k}_3 \mathbf{k}_5})_{\alpha_3 \alpha_5} \\ & + \nu_{\mathbf{k}_1 - \mathbf{k}_4}(\mathcal{P}_{\mathbf{k}_1 \mathbf{k}_5})_{\alpha_1 \alpha_5}(\mathcal{P}_{\mathbf{k}_3 \mathbf{k}_4})_{\alpha_3 \alpha_4}] \\ & \times \left\{ f(\mathbf{k}_2, \alpha_2; \mathbf{k}_5, \alpha_5|t) [n(\mathbf{k}_3, \alpha_3; \mathbf{k}_4, \alpha_4|t) \right. \\ & + \Psi_{\mathbf{k}_3 \alpha_3}^*(t) \Psi_{\mathbf{k}_4 \alpha_4}(t)] \\ & + \frac{1}{2} n^*(\mathbf{k}_2, \alpha_2; \mathbf{k}_3, \alpha_3|t) [f(\mathbf{k}_5, \alpha_5; \mathbf{k}_4, \alpha_4|t) \\ & + \Psi_{\mathbf{k}_5 \alpha_5}(t) \Psi_{\mathbf{k}_4 \alpha_4}(t)] \left. \right\} \\ & + \text{Idem}[(\mathbf{k}_1 \alpha_1) \leftrightarrow (\mathbf{k}_2 \alpha_2)]. \quad (13) \end{aligned}$$

Equations (11)–(13) acquire a simpler form when the state of the system is initially prepared in such a way that at $t=t_0$ only one macrofilled coherent mode $\mathbf{k}=\mathbf{k}_0$ on the lower polariton branch is excited:

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

We can use Eqs. (11)–(13) to find the increments the functions (7) and (10) acquire by time $t=t_0+dt$:

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

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

Now let us substitute

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

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

into the right-hand sides of Eqs. (11)–(13) and again find the increments that the functions (7) and (10) acquire in an infinitesimal time interval dt . The result is

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

i.e., after the quantum fluctuations described by the anomalous distribution function $f(\mathbf{k}_1, \alpha_1; 2\mathbf{k}_0 - \mathbf{k}_1, \alpha_2|t)$, fluctuations characterized by the normal distribution function $n(\mathbf{k}_1, \alpha_1; \mathbf{k}_1, \alpha_2|t)$ are excited in the system.

Substituting

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

into the right-hand sides of Eqs. (11)–(13), we see that the left-hand sides of these equations have the same structure. Thus, with the initial conditions (14) the solution of the system of equations (11)–(13) has the form (15). This result follows solely from momentum conservation in the elementary scattering events of the interacting polaritons.

We can further simplify Eqs. (11)–(13) by keeping in their right-hand sides only resonance terms related solely to states belonging to the lower polariton branch. Allowing for (15) and discarding the rapidly oscillating terms on the right-hand sides of Eqs. (11)–(13), we get

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

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

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

Here

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

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

and

$$\nu_{\text{pol}}(\mathbf{k}, \mathbf{k}_1) = \frac{\nu_{\mathbf{k}-\mathbf{k}_1}}{\sqrt{(1+\Lambda_{\mathbf{k}}^2)(1+\Lambda_{2\mathbf{k}_0-\mathbf{k}}^2)(1+\Lambda_{\mathbf{k}_1}^2)(1+\Lambda_{2\mathbf{k}_0-\mathbf{k}_1}^2)}},$$

$$\tilde{\nu}_{\text{pol}}(\mathbf{k}, \mathbf{k}_1) = \frac{\nu_0 + \nu_{\mathbf{k}-\mathbf{k}_1}}{(1+\Lambda_{\mathbf{k}}^2)(1+\Lambda_{\mathbf{k}_1}^2)} \quad (19)$$

are the polariton–polariton coupling constants. According to (14), the solution of Eqs. (16)–(18) must satisfy the initial condition

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

In what follows we take $t_0 = 0$.

When dealing with equilibrium problems, the quantities of type $\mathcal{F}_{\mathbf{k}}$ or $\tilde{\mathcal{F}}_{\mathbf{k}}$ can be interpreted as order parameters and can be found by solving integral equations.⁹ In the nonequilibrium situation considered here the order parameters are time-dependent, with the result that finding them means solving the system of nonlinear integro-differential equations (16)–(18).

Equations (16)–(18) are invariant under time reversal, i.e., they retain their form when $t \rightarrow -t$, $\Psi_{\mathbf{k}_0} \rightarrow \Psi_{\mathbf{k}_0}^*$, and $f_{\mathbf{k}} \rightarrow f_{\mathbf{k}}^*$.

In addition to additive integrals of the motion, which reflect the conservation of the average values of the number of particles, energy, and momentum of a closed system, Eqs. (16)–(18) are characterized by additional integrals of the motion: $n_{\mathbf{k}}(t) - n_{2\mathbf{k}_0-\mathbf{k}}(t) = \text{const}$ and $|f_{\mathbf{k}}(t)|^2 - n_{\mathbf{k}}(t) \times [1 + n_{\mathbf{k}}(t)] = \text{const}$. Employing the initial conditions (20), we find that

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

This (21) makes it possible to eliminate Eq. (17) from the system of equations (16)–(18).

Clearly, the transformation

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

separates the equation for the phase of the condensate wave function $\Psi_{\mathbf{k}_0}(t)$ from the system of equations (16)–(18). As a result, the system of the evolution equations acquires the form

$$\frac{d}{dt} \mathcal{N}_0(t) = \frac{2}{\hbar} \text{Im} \mathcal{S}_{\mathbf{k}_0}(t) \cdot \mathcal{N}_0(t), \quad (22)$$

$$i\hbar \frac{d}{dt} g_{\mathbf{k}}(t) = \left\{ \hbar(\Omega_{\mathbf{k}} + \Omega_{2\mathbf{k}_0-\mathbf{k}} - 2\Omega_{\mathbf{k}_0}) + [\tilde{\mathcal{F}}_{\mathbf{k}}(t) + \tilde{\mathcal{F}}_{2\mathbf{k}_0-\mathbf{k}}(t) - 2\tilde{\mathcal{F}}_{\mathbf{k}_0}(t)] + [\tilde{\nu}_{\text{pol}}(\mathbf{k}, \mathbf{k}_0) + \tilde{\nu}_{\text{pol}}(2\mathbf{k}_0-\mathbf{k}, \mathbf{k}_0) - 2\nu_{\text{pol}}(\mathbf{k}_0, \mathbf{k}_0)] \times \frac{|\Psi_{\mathbf{k}_0}(0)|^2}{V} \mathcal{N}_0(t) - 2\text{Re} \mathcal{S}_{\mathbf{k}_0}(t) \right\} \times g_{\mathbf{k}}(t) + \sqrt{1 + 4|g_{\mathbf{k}}(t)|^2} \left[\nu_{\text{pol}}(\mathbf{k}, \mathbf{k}_0) \times \frac{|\Psi_{\mathbf{k}_0}(0)|^2}{V} \mathcal{N}_0(t) + \mathcal{F}_{\mathbf{k}}(t) \right], \quad (23)$$

where

$$\mathcal{S}_{\mathbf{k}}(t) = \frac{1}{V} \sum_{\mathbf{k}_1} \nu(\mathbf{k}, \mathbf{k}_1) g_{\mathbf{k}_1}(t) = \mathcal{S}_{2\mathbf{k}_0-\mathbf{k}}(t),$$

and

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

is the relative polariton number density in the condensate.

The steady-state solution of Eqs. (16)–(18),

$$|\Psi_{\mathbf{k}_0}(t)|^2 = \text{const}, \quad n_{\mathbf{k}}(t) \propto \delta(E_{\mathbf{k}} + E_{2\mathbf{k}_0 - \mathbf{k}} - 2E_{\mathbf{k}_0}),$$

$$f_{\mathbf{k}}(t) \propto \delta(E_{\mathbf{k}} + E_{2\mathbf{k}_0 - \mathbf{k}} - 2E_{\mathbf{k}_0}),$$

where $E_{\mathbf{k}}$ is the renormalized polariton energy, was found in Ref. 25. Substitution of this solution into the expression for the total average energy and the total average number of particles suggests that the given solution cannot be realized with the initial condition (20). However, this state of a polariton system can be achieved by irradiating the semiconductor crystal with steady-state laser light.²⁶

4. NUMERICAL RESULTS

The system of equations (22) and (23) is still too complicated even for numerical solution. Hence for the sake of simplicity we ignore the dispersion of the exciton–exciton coupling constant ν . Near an exciton–photon resonance the function $\Lambda_{\mathbf{k}}$ is approximately unity and hence $\tilde{\nu}_{\text{pol}} \approx 2\nu_{\text{pol}} \approx \nu/2$.

The initial condition (20) has the same form for all values of the wave vector \mathbf{k} . Hence Eqs. (22) and (23) imply that in the absence of dispersion in the polariton–polariton coupling constants the \mathbf{k} -dependence of g is achieved by a function of $\Omega_{\mathbf{k}} + \Omega_{2\mathbf{k}_0 - \mathbf{k}} - 2\Omega_{\mathbf{k}_0}$:

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

Using the identity 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 introducing the dimensionless variables

$$T = \lambda t, \quad w_{\mathbf{k}} = \lambda^{-1}(\Omega_{\mathbf{k}} + \Omega_{2\mathbf{k}_0 - \mathbf{k}} - 2\Omega_{\mathbf{k}_0}), \quad (24)$$

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

we can write the system of equations (22) and (23) as follows:

$$\frac{d}{dT} \mathcal{N}_0(T) = 2\text{Im}G(T) \cdot \mathcal{N}_0(T), \quad (25)$$

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

where

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

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

Below, for numerical estimates, we use parameters of polaritons formed by the mixing of photons and 1A-excitons in the ionic crystal of cadmium sulfide (CdS): $\epsilon_B = 9.3$, $\hbar\omega^\perp = 2.55$ eV, $\eta/\hbar = 1.1 \times 10^{14} \text{ s}^{-1}$, $m_\perp = 0.89m_0$ (here m_0 is the free electron mass), and $m_\parallel = 2.85m_0$ (see Ref. 27). We use the model of an isotropic parabolic exciton band with an effective mass $m = (m_\perp^2 m_\parallel)^{1/3}$. The effective Bohr radius of a 1A-exciton in CdS is $a_{\text{ex}} = 28 \text{ \AA}$ (see Ref. 28). The ionization energy is $I_{\text{ex}} = 27$ meV (see Ref. 29). The value of the exciton–exciton coupling constant can be estimated by the formula $\nu \equiv \nu_0 = (26\pi/3) I_{\text{ex}} a_{\text{ex}}^3$ (see Refs. 4 and 30) and amounts to $4.3 \times 10^{-33} \text{ erg} \cdot \text{cm}^3$. The interaction of excitons with acoustic phonons in CdS is extremely weak. Only the interaction with a single branch of optical phonons with an energy $\hbar\omega_{\text{ph}} \sim 38$ meV, which are characterized by considerable dispersion, can play a noticeable role.³¹

It the initial evolution of the system, when a considerable fraction of polaritons is still in the condensate and the number of extracondensate polaritons is fairly low, we can ignore the terms with the function $G(T)$ in Eqs. (25) and (26). The solution of such a “truncated” system of equations has the form

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

(here the effect of the small fraction of extracondensate polaritons on the condensate is ignored),

$$n(w, T)$$

$$\begin{aligned} &= \begin{cases} -\frac{1}{w(w+2)} \sinh^2 \left[\frac{T}{2} \sqrt{-w(w+2)} \right] & \text{if } -2 \leq w \leq 0, \\ \frac{1}{w(w+2)} \sinh^2 \left[\frac{T}{2} \sqrt{w(w+2)} \right] & \text{if } w < -2 \text{ or } w > 0. \end{cases} \end{aligned} \quad (28)$$

According to (28), in the initial stage of evolution there is intense excitation of extracondensate polaritons in the range $-2 \leq w \leq 0$, where the distribution function $n(w)$ is shaped like a symmetric bell with the maximum at $w = -1$. As we move away from this range, the distribution function decreases in an oscillatory manner.

The numerical solution of the system of equations (25) and (26) is done in two stages. First we find the explicit form of the ρ vs w dependence for various values of the condensate density $|\Psi_{\mathbf{k}_0}(0)|^2/V$ and of the wave vector \mathbf{k}_0 . Then this dependence is used to integrate the evolution equations (25) and (26). The results of integration for $|\mathbf{k}_0| = 3.6 \times 10^5 \text{ cm}^{-1}$ are depicted in Figs. 1–3. This choice of the value of $|\mathbf{k}_0|$ corresponds to an exciton–photon resonance lying slightly below the bottom of the exciton band.

Disintegration of the nonequilibrium polariton condensate is described by the curve in Fig. 1. Up to the moment

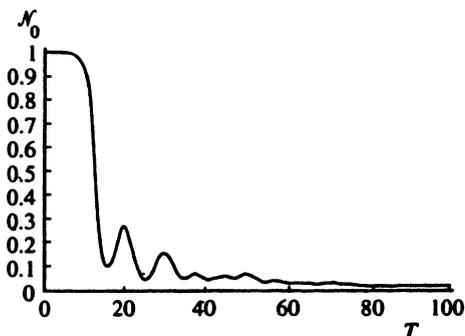


FIG. 1. The relative number density of polaritons in the condensate as a function of the dimensionless time T .

$T \approx 7$ of dimensionless time the depletion of the condensate proceeds rather slowly. Then there is a sharp drop, as a result of which by $T \approx 15$ only 10% of the original polaritons remain in the condensate. Further disintegration of the condensate is accompanied by nutation oscillations, in the course of which the condensate density is partially restored (up to 30% at $T \approx 20$). Nutation oscillations are absent if in Eq. (26) we ignore the function $G(T)$, which describes the combined effect of all pairs of extracondensate polaritons with the same value of total momentum $2\hbar\mathbf{k}_0$ per pair. Thus, in the disintegration process the condensate oscillations are related to the correlation of the states of separate pairs of extracondensate polaritons. By $T \approx 50$ the amplitude of the oscillations and the disintegration rate noticeably diminish. These results cannot be obtained by introducing phenomenological constants into the dynamical equation for the condensate's wave function.

Figure 2 depicts the distribution function of extracondensate polaritons at moments chosen within the interval in which the polariton condensate undergoes the most rapid and substantial variations. The curve corresponding to $T=10$ is shaped like a symmetric bell with its maximum at $w = -1$, just as the curve describing the function (28). By $T=15$ the distribution function increases considerably in magnitude, while remaining symmetrical, and the maximum moves toward higher energies of extracondensate polaritons. The increase in the number of extracondensate polaritons by a factor greater than 10 is consistent with the fact that the function

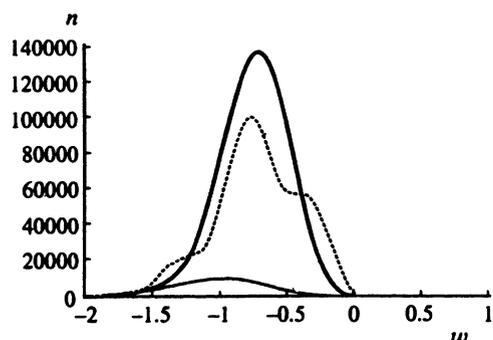


FIG. 2. The distribution function of extracondensate polaritons at $T=10$ (solid light curve), 15 (solid heavy curve), and 20 (dotted curve).

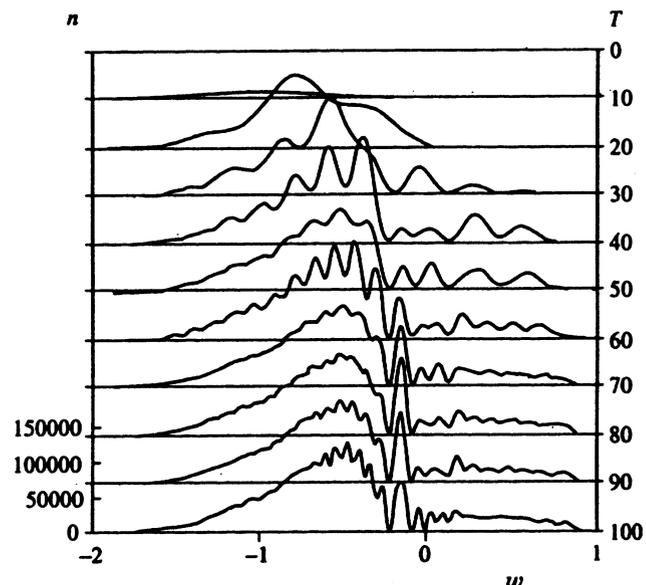


FIG. 3. The distribution function of extracondensate polaritons at different moments of the dimensionless time T . The marks on the left vertical axis give an idea of the scale of the function.

$\mathcal{N}_0(T)$ has a local minimum at $T \approx 15$. At $T=20$ the envelope of the distribution current becomes distorted and the area under it decreases. The latter is consistent with the fact that the function $\mathcal{N}_0(T)$ has a local maximum at $T \approx 20$. Thus, the distribution function rises and falls in step with the oscillations of the condensate's density.

The subsequent evolution of the distribution function is depicted in Fig. 3. By $T=30$ the envelope acquires oscillations which emerge not only in the dependence of the distribution function on the parameter w but also in the dependence on the dimensionless time T (unfortunately, we were unable to reflect this fact in Fig. 3). This means that if, for instance, the distribution function at $T \approx 40$ has a local maximum at $w = -1$, at another time it has a local minimum at the same point. With the passage of time the frequency oscillations become denser and shallower. This results in chaotic ripples appearing on the smooth envelope of the distribution function.

At $T \approx 30$ the distribution function acquires a short-wave wing. By $T=100$ the distribution function is localized in the range $-2 < w < 1$. Of special interest here is the ignition, followed by partial extinction, of an additional sharp peak localized in the range $-0.22 < w < -0.08$ with its maximum at $w \approx -0.15$. The time interval in which the additional peak is formed and exists is characterized by low values of the condensate density and slow variations of this density (see Figs. 1 and 3). The position of the additional peak does not change with the passage of time, and the peak reaches its maximum height at $T \approx 80$.

Changes in the initial condensate density $|\Psi_{\mathbf{k}_0}(0)|^2/V$ result in variations in only the scale of the dependencies discussed here because of the corresponding transformation of the dimensionless arguments (24). This is a corollary of the extremely weak dependence of the solution of Eqs. (25) and (26) on the initial condensate density via the function

$\rho(w)$ contained in the definition of $G(T)$. According to Figs. 2 and 3, $g(w, T)$ is nonzero only in the interval $-2 < w < 1$. Numerical calculations show that in this interval $\rho(w)$ is essentially independent of w because of the specific features of the dispersion of the lower polariton branch near an exciton–photon resonance. This makes it possible to replace $\rho(w)$ with $\rho(0)$ in the definition (27) of $G(T)$. As a result we have

$$G(T) \approx \rho(0) \int dw g(w, T).$$

But according to (27) and (24), $\rho(0)$ is independent of the initial density of the condensate and for fixed crystal characteristics is determined exclusively by the size of the wave vector \mathbf{k}_0 . At $|\mathbf{k}_0| = 3.6 \times 10^5 \text{ cm}^{-1}$ we have $\rho(0) \sim 6.8 \times 10^{-6}$.

Note that although Eqs. (16)–(18) are reversible in time, their solution is not.³²

5. CONCLUSION

Let us take a specific physical situation in which the above physical phenomena might show up. Suppose that a semiconducting crystal of thickness L in a ring cavity is irradiated by a pulse of resonant laser light of length τ . The ring cavity is needed to confine the condensate and the extracondensate excitations to the system. If the wave vectors of the condensate and scattered modes satisfy $kL \gg 1$, the propagation of the laser light in the resonant medium should be interpreted as the propagation of interacting mixed exciton–photon modes, or polaritons. The polaritons are formed in a time interval $\tau_{\text{pol}} \sim \hbar/2\eta \ll \tau$ as a result of interconversion of the photons of the internal electromagnetic field (the field inside the semiconducting sample) and excitons with the same value of the wave vector. For a 1A-exciton in a CdS crystal, $\tau_{\text{pol}} \approx 7$ fs.

We assume that the length of the excitation pulse is limited by the inequality $\tau \ll \tau_* = 10\lambda^{-1}$, which implies that in the course of τ the polariton condensate has no time to “feel” the effect of the scattered modes and to be noticeably depleted (see Fig. 1). Then in the above-mentioned interval the propagation of the condensate can be described by classical equations: the linear inhomogeneous Maxwell equation for the potential of the electromagnetic field and the nonlinear nonhomogeneous Schrödinger equation for the exciton amplitude.^{1,4} The inhomogeneous terms in the equations are related to the exciton–photon interaction. Integrating the Maxwell equation with allowance for the conditions at the crystal–vacuum interface, we find that the internal electromagnetic field is a linear combination of (1) the secondary field generated by exciton polarization, (2) the field whose source is the crystal’s boundary (this field emerges as a result of the action of external radiation on the boundary²⁹), and (3) the field that existed in the crystal in the infinitely remote past (as $t \rightarrow -\infty$). The latter component must be identically zero for the short excitation pulse considered here. Thus it can be said that after time τ has elapsed the crystal contains a condensate polariton wave generated by a source at the crystal’s boundary. The role of the excitation pulse consists

in “creating” the initial condition that the condensate wave function must obey at the instant the pulse ceases to exist.

The above line of reasoning has the following drawback. The use of a laser pulse of finite length leads to excitation of a polariton condensate in a finite spectral range of width $\Delta\Omega \sim \tau^{-1}$, which contradicts the initial condition (14). This contradiction, however, is unimportant if $\Delta\Omega \ll \lambda$, i.e., when $\tau \gg 0.1\tau_*$. For an initial polariton number density in the CdS crystal of $10^{16} - 10^{18} \text{ cm}^{-3}$ we have 80–0.8ps, respectively.

Thus, the length of the excitation pulse must meet fairly stringent and somewhat contradictory conditions: $0.1\tau_* \ll \tau < \tau_*$. Nevertheless, it can be expected that at least some features of the temporal evolution of the system of initially coherent polaritons uncovered within the framework of the stated model problem will show up, even if the first condition is not met or if the second is slightly violated.

The source of the laser light exciting the coherent polariton wave in the crystal must be sufficiently powerful that the nutation oscillations of the condensate density and the specific features of the distribution function of the scattered modes have time to appear before relaxation processes involving optical phonons set in or photons escape from the cavity. The physical phenomena discussed in Sec. 4 occur in a time interval $1.5\tau_* < \tau < 10\tau_*$ (see Figs. 1–3). We assume, for the sake of definiteness, that the minimum relaxation time τ_r of condensate or scattered polaritons on phonons does not exceed the photon lifetime in the cavity, which is related to the transmissivity of the cavity’s mirrors. By varying the frequency ω of the resonant radiation impinging on the crystal over a certain spectral range lying below the exciton transition frequency ω^1 , a situation can be created in which, on the one hand, the kinematic relationships can be made to allow for real two-quantum escape of polaritons from the condensate and, on the other, the condition $\mathcal{T} < \tau_r$ can be made to hold (here $\mathcal{T} = (1.5 - 10)\tau_*$ is the delay time determined by the moment of observation). The time τ_r strongly depends on the state of the polaritons in the band.^{24,29} At moderate temperatures ($k_B T \ll \hbar\omega_{\text{ph}}$, with k_B the Boltzmann constant), when anti-Stokes scattering of polaritons by optical photons is negligible, and the polariton states with energies close to the bottom of the exciton band are characterized by extremely long relaxation times ($\tau_r \sim 0.1 \mu\text{s} - 1$ ns; see Ref. 24). This corresponds to a sharp increase in the density of the possible final states in polariton–photon scattering, when states are transformed from exciton-like into photon-like.

Observing the disintegration process requires the thickness of the semiconducting sample to be bounded by $L < v_g \mathcal{T}$, where v_g is the polariton group velocity, which depends on the choice of wave vector \mathbf{k}_0 (see Ref. 25). The value $v_g = 9.4 \times 10^9 \text{ cm} \cdot \text{s}^{-1}$ corresponds to $|\mathbf{k}_0| = 3.6 \times 10^5 \text{ cm}^{-1}$. We also note that one of the modes emerging in an elementary scattering event lies in the photon-like section of the polariton branch and has a component of the wave vector directed opposite to the direction of propagation of the excitation pulse.⁵ This mode rapidly leaves the crystal. For it to return to the scattering region in a time interval much shorter than τ_* , the thickness L of the semiconducting sample must be much smaller than $(c/\sqrt{\epsilon_B})\tau_*$.

The authors are grateful to A. L. Ivanov and P. I. Khadzhi for interest in this work.

- ¹S. A. Moskalenko, P. I. Khadzhi, and A. Kh. Rotaru, *Solitons and Nutation in the Excitonic Part of the Spectrum* [in Russian], Shtiintsa, Kishinev (1980).
- ²A. L. Ivanov and L. V. Keldysh, *Zh. Éksp. Teor. Fiz.* **84**, 404 (1983) [Sov. Phys. JETP **57**, 234 (1983)].
- ³M. V. Lebedev, *Zh. Éksp. Teor. Fiz.* **101**, 957 (1992) [Sov. Phys. JETP **74**, 513 (1992)].
- ⁴S. A. Moskalenko, *Introduction to the Theory of High-Density Excitons* [in Russian], Shtiintsa, Kishinev (1983).
- ⁵M. I. Shmiglyuk and V. N. Pitei, *Coherent Polaritons in Semiconductors* [in Russian], Shtiintsa, Kishinev (1989).
- ⁶V. N. Pitei, M. I. Shmiglyuk, V. T. Zyukov *et al.*, in *Coherent States and Phase Transitions in a System of High-Density Polaritons* [in Russian], Shtiintsa, Kishinev (1985).
- ⁷N. N. Bogolyubov, *Vestnik Moskov. Gos. Univ.* **7**, 43 (1947); N. N. Bogolyubov, *Selected Works*, Vol. 2 [in Russian], Naukova Dumka, Kiev (1970), p. 242.
- ⁸L. D. Landau and E. M. Lifshitz, *Quantum Mechanics: Non-relativistic Theory*, 3rd ed., Pergamon Press, Oxford (1977).
- ⁹J. R. Schrieffer, *Theory of Superconductivity*, W. A. Benjamin, New York (1964).
- ¹⁰N. N. Bogolyubov, Preprint JINR D-788, Dubna (1961); N. N. Bogolyubov, *Selected Works*, Vol. 3 [in Russian], Naukova Dumka, Kiev (1971), p. 174.
- ¹¹N. N. Bogolyubov, Jr., and B. I. Sadovnikov, *Several Aspects of Statistical Mechanics*, [in Russian], Vysshaya shkola, Moscow (1975).
- ¹²V. N. Popov and V. S. Yarunin, *Collective Effects in the Quantum Statistics of Radiation and Matter*, Leningrad State Univ. Press, Leningrad (1985) [in Russian].
- ¹³S. A. Moskalenko, A. H. Rotaru, and Yu. M. Shvera, in *Laser Optics of Condensed Matter*, J. L. Birman, H. Z. Cummins, and A. A. Kaplyanski (eds.), Plenum Press, New York (1988), p. 331.
- ¹⁴V. R. Misko, S. A. Moskalenko, A. H. Rotaru *et al.*, *Phys. Status Solidi B* **159**, 477 (1990).
- ¹⁵I. V. Belousov and Yu. M. Shvera, *Phys. Status Solidi B* **159**, 91 (1990).
- ¹⁶I. V. Belousov and Yu. M. Shvera, *Z. Phys. B* **90**, 51 (1993).
- ¹⁷L. V. Keldysh, *Zh. Éksp. Teor. Fiz.* **47**, 1515 (1964) [Sov. Phys. JETP **20**, 1018 (1965)].
- ¹⁸I. V. Belousov and Yu. M. Shvera, *Teoret. Mat. Fiz.* **85**, 237 (1990).
- ¹⁹D. N. Zubarev, *Nonequilibrium Statistical Thermodynamics*, Consultants Bureau, New York (1974).
- ²⁰I. V. Belousov and Yu. M. Shvera, in *Excitons and Biexcitons in Dimension-Limited Systems* [in Russian], Shtiintsa, Kishinev (1990), p. 133.
- ²¹I. V. Belousov and Yu. M. Shvera, in *Interaction of Excitons and Laser Light*, Shtiintsa [in Russian], Kishinev (1991) p. 57.
- ²²L. V. Keldysh, in *Problems of Theoretical Physics* [in Russian], Nauka, Moscow (1972), p. 433.
- ²³I. A. Akhiezer and S. V. Peletminskiĭ, *Methods of Statistical Physics*, Pergamon Press, Oxford (1980).
- ²⁴M. S. Brodin, É. N. Myasnikov, and S. V. Marisova, *Polaritons in Crystal Optics* [in Russian], Naukova Dumka, Kiev (1984).
- ²⁵I. V. Belousov and Yu. M. Shvera, in *Bose–Einstein Condensation*, A. Griffin, D. W. Snoke, and S. Stringari (eds.), Cambridge Univ. Press, New York (1995), p. 513.
- ²⁶V. L. Safonov, *Physica A* **188**, 675 (1992).
- ²⁷É. S. Koteles, in: *Excitons*, Nauka, Moscow (1985), p. 68.
- ²⁸B. S. Wang and J. L. Birman, *Phys. Rev. B* **42**, 9609 (1990).
- ²⁹W. F. Brinkman, T. M. Rice, and B. Bell, *Phys. Rev. B* **8**, 1570 (1966).
- ³⁰A. I. Bobrysheva, M. F. Miglei, and M. I. Shmiglyuk, *Phys. Status Solidi B* **53**, 71 (1972).
- ³¹A. S. Davydov, *Solid-State Theory* [in Russian], Nauka, Moscow (1976).
- ³²G. M. Zaslavskii, (Zaslavsky), *Chaos in Dynamical Systems*, Harwood, Chur (1985).

Translated by Eugene Yankovsky