Quantum kinetic equations for interacting bosons and their application for polariton parametric oscillators

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We derive quantum kinetic equations describing a system of interacting polaritons relaxing the Markovian approximation. We demonstrate the crucial role played by the nonclassical four-particle correlators in the dynamics. The decoherence process is shown to be responsible for the transition from the quantum limit to the classical limit. The general formalism is applied to the description of the dynamics of the polariton parametric oscillator. It allows accounting self-consistently for both signal-idler correlations and pump depletion.

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I. INTRODUCTION

The systems of interacting bosons are the focus of experimental and theoretical research at present. The interest in these systems is stimulated by recent experimental demonstration of Bose Einstein condensation (BEC) of cold atoms[1–3] and exciton-polaritons in microcavities.[4] BEC of cold alkali atoms is characterized by extremely low critical temperatures (in nanokelvin region), while exciton-polaritons can be condensed up to room temperature, in theory.[5] On the other hand, strictly speaking, BEC is forbidden in two-dimensional systems like planar microcavities, so that the observed phenomena[4,6] are related to the quasicondensation of the polaritons in a finite size system. In an ideal infinite planar microcavity there is no BEC, but a superfluid phase transition may take place,[7] which obviously requires polariton-polariton interactions.

The dynamics of condensation of interacting bosons is an extremely complex process which is successfully treated theoretically only in particular cases and using strong approximations. In such bosonic systems as cold atoms or conventional superfluids, the static models using thermodynamic limit provide quite reliable results. On the other hand, exciton-polaritons are always out of thermal equilibrium due to their finite lifetime. Successful attempts to account for the polariton lifetime introducing some effective chemical potential have been done recently.[8–10] However, an important class of experiments done at resonant optical excitation of exciton-polaritons requires theoretical treatment beyond the thermal equilibrium assumption. This concerns especially the description of polariton parametric oscillators (PPOs) and amplifiers.[11–17] Here, we present a dynamical quantum model allowing the description of various systems of interacting bosons and apply it to the microcavity parametric oscillators. Our model goes beyond existing theories as it relaxes the Born-Markov approximation. It allows describing the transition between the quantum and classical limits for the microcavity PPOs.

To be specific, we shall consider a system of n_D interacting excitons and assume that the following condition is satisfied:

$$n_D a_B^D \ll 1, \qquad (1)$$

where D is the dimensionality of the system ($D=1,2,3$) and a_B is the exciton Bohr radius. We shall further assume that this excitonic system is strongly coupled with light in a semiconductor microcavity, so that mixed exciton-photon quasiparticles called exciton polaritons are formed. Being combinations of quantum well excitons and cavity photons, the cavity polaritons retain the properties of both. The presence of the photonic component results in the extremely small effective mass of cavity polaritons (10^{-4}–10^{-5} of the electron mass),[5] while the excitonic component makes possible effective polariton-phonon and polariton-polariton interactions. These factors are crucial for polariton BEC, whose critical temperature was predicted to be extremely high (tens of kelvins for GaAs and CdTe microcavities, up to room temperatures for GaN and ZnO cavities[5]). Recently, BEC of polaritons has been observed experimentally in a CdTe microcavity at about 20 K.[4] Similar to the exciton condensate as studied theoretically by Keldysh and Kopaev 42 years ago,[18] polariton condensate emits coherent light and, thus, can be used for the creation of a new generation optoelectronic device known as "polariton laser."[19]

Investigation of the mechanisms of polariton redistribution (and bosons, in general) in the reciprocal space is crucial for the comprehension of the formation of BECs. For the cavity polaritons, two mechanisms are of major importance: the polariton-phonon and polariton-polariton interactions. The former is dominant at small densities, while the latter becomes dominant in the nonlinear regime and especially at the bottleneck region, where polariton relaxation with acoustic phonons is no more efficient.[20] Polariton-polariton scattering is even more important in the case of resonant optical pumping that creates coherent macroscopic population of polaritons at their lower dispersion branch. In this case, two main nonlinear mechanisms have been identified, which are polariton parametric scattering[13,14,17] and the blueshift of the polariton dispersion.[6] These two mechanisms often occur simultaneously, leading to the number of intriguing nonlinear phenomena such as bistability of the polariton system.[21–24]

The existing models of the PPO either consider all three states involved in the parametric process as classical fields coupled by a four-wave mixing process[24] or consider the case of cw pumping, neglecting the pump depletion.[13,14] The exception is the recent work of Glazov and Kavokin,[25] where the hyperspin formalism was applied for the analysis of the parametric amplifier. The hyperspin formalism allows describing up to a certain point the quantum correlations in a three-level system, while its extension to more complex systems would require extremely heavy analytics. Other works[22,23] are based on Gross-Pitaevskii equations. This allows taking into account all states of reciprocal space, but the decoherence or relaxation associated with phonons are completely neglected as well as processes of spontaneous polariton-polariton scattering.

In the present paper, we derive quantum kinetic equations for the system of interacting bosons. They describe the dynamics of the occupation numbers and of nonclassical off-diagonal four-particle correlators. We argue that decoherence process leading to the decay of the nonclassical correlators leads to the transition between the quantum oscillatory regime and semiclassical relaxation regime. We show that in the limit of very fast dephasing, the system of quantum equations becomes similar to the semiclassical system of Boltzmann equations. These quantum kinetic equations are then applied to describe the three state model corresponding to the PPO.

The paper is organized as follows. In Sec. II, we present the model and discuss the proposed set of kinetic equations for the occupation numbers and nonclassical correlators. In Sec. III, we discuss the mechanisms of decoherence and study the transition from quantum limit to the classical limit. In Sec. IV, we apply the developed formalism to a PPO. Section V contains the conclusions.

II. QUANTUM KINETIC EQUATION FOR A SYSTEM OF INTERACTING BOSONS

Here and further, we consider a system of spinless interacting bosons, e.g., cavity polaritons. We address the readers interested in the spin dynamics of exciton-polaritons to the recent review paper.[26] Here, we consider a model quantum system described by the Hamiltonian

$$H=\sum_{\mathbf{k}}\varepsilon_{\mathbf{k}}a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}+\sum_{\mathbf{k},\mathbf{q}}U_{\mathbf{k},\mathbf{q}}a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}-\mathbf{q}}(b_{\mathbf{q}}+b_{-\mathbf{q}}^{\dagger})+\frac{1}{2}\sum_{\mathbf{k},\mathbf{k}',\mathbf{q}}V_{\mathbf{k},\mathbf{k}',\mathbf{q}}a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}'}^{\dagger}a_{\mathbf{k}-\mathbf{q}}a_{\mathbf{k}'+\mathbf{q}},\tag{2}$$

where operators $a_{\mathbf{k}}$ are the boson annihilation operators for polaritons and $b_{\mathbf{q}}$ the acoustic phonon annihilation operators. The first term corresponds to the free particle motion, the second term describes exciton-acoustic phonon scattering, and the third term describes polariton-polariton scattering. The latter arises from the Coulomb interaction between the excitonic fractions of two colliding polaritons and plays a major role in polariton relaxation. The matrix element of this scattering is determined by the exciton binding energy E_B, exciton Bohr radius a_B, and the area occupied by the condensate S. Roughly, it can be estimated as

$$V_{\mathbf{k},\mathbf{k}';\mathbf{k}'',\mathbf{k}'''}=\langle\mathbf{k},\mathbf{k}'|V_{\mathrm{int}}|\mathbf{k}'',\mathbf{k}'''\rangle\sim\frac{E_Ba_B^2}{S}X_{\mathbf{k}}^{*}X_{\mathbf{k}'}^{*}X_{\mathbf{k}''}X_{\mathbf{k}'''}\delta_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''-\mathbf{k}'''},\tag{3}$$

where $X_{\mathbf{k}}$ is a Hopfield coefficient giving the percentage of the excitonic fraction in the state $\mathbf{k}$, and the delta function ensures the momentum conservation during the scattering act. An estimation of this quantity within the mean-field approximation[27] has given a prefactor of 6 in the left hand side of Eq. (3). We neglected in Eq. (3) the saturation terms,[28] assuming that condition (1) is satisfied.

When considering the dynamics of the polariton system described by Eq. (2), the phonon field can be treated classically. The usual way to deal with it is to use the Born-Markov approximation for the Liouville–von Neumann equation for the density matrix of the system. The resulting system of kinetic equations is of the Boltzmann type. This procedure is well described in literature (see, e.g., Ref. 29) and we will not further consider the interaction with acoustic phonons in the rest of the paper. We note, however, that a strong advantage of the approach we use, with respect to the models assuming the full coherence as the Gross-Pitaevskii equations, is that it allows taking simultaneously into account the coherent and noncoherent aspects of the polariton dynamics.

Let us now consider the term describing particle-particle interactions [last term in Eq. (3)]. Formally, the Born-Markov approximation can be applied also in this case.[30] The justification of this approximation is, however, less straightforward since there is no classical reservoir in the system. To consider the dynamics of the system, we start from the Liouville–von Neumann equation which reads

$$i\hbar\frac{d\rho}{dt}=[H;\rho]=\sum_{\mathbf{k}}\varepsilon_{\mathbf{k}}[a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}\rho-\rho a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}]+\frac{1}{2}\sum_{\mathbf{k},\mathbf{q}}V_{\mathbf{k},\mathbf{k}',\mathbf{q}}[a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}'}^{\dagger}a_{\mathbf{k}-\mathbf{q}}a_{\mathbf{k}'+\mathbf{q}}\rho-\rho a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}'}^{\dagger}a_{\mathbf{k}-\mathbf{q}}a_{\mathbf{k}'+\mathbf{q}}].\tag{4}$$

It yields the following dynamics of the occupation numbers $N_{\mathbf{k}}=\mathrm{Tr}(a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}\rho)$:

$$\frac{dN_{\mathbf{k}}}{dt}=\mathrm{Tr}\left(a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}\frac{d\rho}{dt}\right)=-\frac{1}{\hbar}\sum_{k',q}\mathrm{Im}[V_{\mathbf{k},\mathbf{k}',\mathbf{q}}\langle a_{\mathbf{k}-\mathbf{q}}^{\dagger}a_{\mathbf{k}'+\mathbf{q}}^{\dagger}a_{\mathbf{k}}a_{\mathbf{k}'}\rangle]=-\frac{1}{\hbar}\sum_{k',q}\mathrm{Im}[V_{\mathbf{k},\mathbf{k}',\mathbf{q}}A_{\mathbf{k},\mathbf{k}',\mathbf{q}}].\tag{5}$$

The right part of Eq. (5) contains the fourth-order correlators $A_{\mathbf{k},\mathbf{k}',\mathbf{q}}=\langle a_{\mathbf{k}-\mathbf{q}}^{\dagger}a_{\mathbf{k}'+\mathbf{q}}^{\dagger}a_{\mathbf{k}}a_{\mathbf{k}'}\rangle=\mathrm{Tr}[\rho a_{\mathbf{k}-\mathbf{q}}^{\dagger}a_{\mathbf{k}'+\mathbf{q}}^{\dagger}a_{\mathbf{k}}a_{\mathbf{k}'}]$. It follows from Eq. (5) that the total number of particles in the system is conserved, $(d/dt)\Sigma_{\mathbf{k}}N_{\mathbf{k}}=0$, as it should be in the absence of damping. Note that Eq. (5) is obtained from Eq. (4) without any simplifying assumptions. In order to take into account the finite lifetime of exciton-polaritons, an additional term $-N_{\mathbf{k}}/\tau_{\mathbf{k}}$ should be introduced into Eq. (5).

To complete the set of kinetic equations, we derive an expression for the temporal derivative of $A_{\mathbf{k},\mathbf{k}',\mathbf{q}}$ which reads

$$\begin{aligned}\frac{dA_{\mathbf{k},\mathbf{k}',\mathbf{q}}}{dt} &= \mathrm{Tr}\left(\frac{d\rho}{dt}a_{\mathbf{k}}a_{\mathbf{k}'}a^{\dagger}_{\mathbf{k}-\mathbf{q}}a^{\dagger}_{\mathbf{k}'+\mathbf{q}}\right)\\ &= \frac{i}{\hbar}\mathrm{Tr}([\rho;H]a^{\dagger}_{\mathbf{k}-\mathbf{q}}a^{\dagger}_{\mathbf{k}'+\mathbf{q}}a_{\mathbf{k}}a_{\mathbf{k}'})\\ &= \frac{i}{\hbar}(\varepsilon_{\mathbf{k}'+\mathbf{q}}+\varepsilon_{\mathbf{k}-\mathbf{q}}-\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{k}'})A_{\mathbf{k},\mathbf{k}',\mathbf{q}}\\ &\quad+\frac{i}{\hbar}\sum_{\mathbf{k}'',\mathbf{q}'}[V_{\mathbf{k},\mathbf{k}'',\mathbf{q}'}\langle a_{\mathbf{k}''}a^{\dagger}_{\mathbf{k}-\mathbf{q}'}a^{\dagger}_{\mathbf{k}''+\mathbf{q}'}a^{\dagger}_{\mathbf{k}'}a_{\mathbf{k}-\mathbf{q}}a_{\mathbf{k}'+\mathbf{q}}\rangle\\ &\quad+V_{\mathbf{k}',\mathbf{k}'',\mathbf{q}'}\langle a^{\dagger}_{\mathbf{k}}a_{\mathbf{k}''}a^{\dagger}_{\mathbf{k}'-\mathbf{q}'}a^{\dagger}_{\mathbf{k}''+\mathbf{q}'}a_{\mathbf{k}-\mathbf{q}}a_{\mathbf{k}'+\mathbf{q}}\rangle\\ &\quad-V_{\mathbf{k}-\mathbf{q},\mathbf{k}'',\mathbf{q}'}\langle a^{\dagger}_{\mathbf{k}}a^{\dagger}_{\mathbf{k}'}a^{\dagger}_{\mathbf{k}''}a_{\mathbf{k}-\mathbf{q}-\mathbf{q}'}a_{\mathbf{k}''+\mathbf{q}'}a_{\mathbf{k}'+\mathbf{q}}\rangle\\ &\quad-V_{\mathbf{k}'+\mathbf{q},\mathbf{k}'',\mathbf{q}'}\langle a^{\dagger}_{\mathbf{k}}a^{\dagger}_{\mathbf{k}'}a_{\mathbf{k}-\mathbf{q}}a^{\dagger}_{\mathbf{k}''}a_{\mathbf{k}'+\mathbf{q}-\mathbf{q}'}a_{\mathbf{k}''+\mathbf{q}'}\rangle]\\ &= \frac{i}{\hbar}(\varepsilon_{\mathbf{k}'+\mathbf{q}}+\varepsilon_{\mathbf{k}-\mathbf{q}}-\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{k}'})A_{\mathbf{k},\mathbf{k}',\mathbf{q}}\\ &\quad+F_1+F_2+F_3+F_4. \end{aligned}\tag{6}$$

In Eq. (6), we have expanded the sum of the sixth-order correlators into four parts, $F_1+F_2+F_3+F_4$. At this stage, we are going to break the hierarchy decoupling the sixth-order correlators. Our goal is to write a closed system of equations for the occupation numbers and the fourth-order correlators $A_{\mathbf{k},\mathbf{k}',\mathbf{q}}$.

Depending on the wave vectors, all the terms in Eq. (6) can be collected into four distinct groups

(1) The term F_1 corresponds to the correlators with $\mathbf{q}'=0$. It describes forward scattering and reads

$$\begin{aligned}F_1 &= \frac{i}{\hbar}\sum_{\mathbf{k}''}(V_{\mathbf{k}-\mathbf{q},\mathbf{k}'',0}+V_{\mathbf{k}'+\mathbf{q},\mathbf{k}'',0}-V_{\mathbf{k},\mathbf{k}'',0}-V_{\mathbf{k}',\mathbf{k}'',0})\\ &\quad\times\langle a^{\dagger}_{\mathbf{k}'+\mathbf{q}}a^{\dagger}_{\mathbf{k}-\mathbf{q}}a_{\mathbf{k}}a_{\mathbf{k}'}a^{\dagger}_{\mathbf{k}''}a_{\mathbf{k}''}\rangle\\ &\approx \frac{i}{\hbar}A_{\mathbf{k},\mathbf{k}',\mathbf{q}}\sum_{\mathbf{k}''}(V_{\mathbf{k}-\mathbf{q},\mathbf{k}'',0}+V_{\mathbf{k}'+\mathbf{q},\mathbf{k}'',0}\\ &\quad-V_{\mathbf{k},\mathbf{k}'',0}-V_{\mathbf{k}',\mathbf{k}'',0})N_{\mathbf{k}''},\end{aligned}\tag{7}$$

where in the passage from the first to the second line we have used the mean-field approximation, i.e., we neglected the correlations between the states inside and outside the correlator. The term F_1 is responsible for the energy renormalization of the states coupled by polariton-polariton interactions.

(2) The second term F_2 contains the correlators, in which the momenta of the three incoming creation operators coincide with momenta of the three annihilation operators. It accounts for the scattering of the states forming the correlator. As we shall argue below, it is the most important correlator for the polariton-polariton interactions in the limit of strong dephasing. We have

$$\begin{aligned}F_2 &= \frac{i}{\hbar}V_{\mathbf{k},\mathbf{k}',\mathbf{q}}[\langle a^{\dagger}_{\mathbf{k}}a_{\mathbf{k}}a^{\dagger}_{\mathbf{k}'}a_{\mathbf{k}'}a_{\mathbf{k}'+\mathbf{q}}a^{\dagger}_{\mathbf{k}'+\mathbf{q}}\rangle+\langle a^{\dagger}_{\mathbf{k}}a_{\mathbf{k}}a^{\dagger}_{\mathbf{k}'}a_{\mathbf{k}'}a^{\dagger}_{\mathbf{k}-\mathbf{q}}a_{\mathbf{k}-\mathbf{q}}\rangle\\ &\quad-\langle a^{\dagger}_{\mathbf{k}'}a_{\mathbf{k}'}a^{\dagger}_{\mathbf{k}-\mathbf{q}}a_{\mathbf{k}-\mathbf{q}}a^{\dagger}_{\mathbf{k}'+\mathbf{q}}a_{\mathbf{k}'+\mathbf{q}}\rangle\\ &\quad-\langle a_{\mathbf{k}}a^{\dagger}_{\mathbf{k}}a^{\dagger}_{\mathbf{k}-\mathbf{q}}a_{\mathbf{k}-\mathbf{q}}a^{\dagger}_{\mathbf{k}'+\mathbf{q}}a_{\mathbf{k}'+\mathbf{q}}\rangle]\\ &\approx \frac{i}{\hbar}V_{\mathbf{k},\mathbf{k}',\mathbf{q}}[N_{\mathbf{k}}N_{\mathbf{k}'}(N_{\mathbf{k}-\mathbf{q}}+N_{\mathbf{k}'+\mathbf{q}}+1)\\ &\quad-N_{\mathbf{k}-\mathbf{q}}N_{\mathbf{k}'+\mathbf{q}}(N_{\mathbf{k}}+N_{\mathbf{k}'}+1)],\end{aligned}\tag{8}$$

where we used the symmetry of the matrix element $V_{\mathbf{k},\mathbf{k}',\mathbf{q}}=V_{\mathbf{k}',\mathbf{k},-\mathbf{q}}=V_{\mathbf{k}-\mathbf{q},\mathbf{k}'+\mathbf{q},-\mathbf{q}}=V_{\mathbf{k}'+\mathbf{q},\mathbf{k}-\mathbf{q},\mathbf{q}}$ for the systems with time inversion and again used the mean-field approximation to pass from the first to the second line.

Equation (8) has a clear physical sense. The term in its right hand side is the collision term of the classical Boltzmann equation (note, however, that the Boltzmann equation does not contain fourth-order correlators). We see that within this assumption the dynamics of the fourth-order correlator is governed by the dynamics of the occupation numbers. The term F_2 provides the spontaneous buildup of correlators, which initially were absent in the system, and also takes into account the effects of the final state bosonic stimulation.

(3) The term F_3 contains the correlators, in which the momentum of one creation operator coincides with the momentum of one annihilation operator. These correlators are decoupled following the usual procedure used to decouple the Bogoliubov chains,

$$\begin{aligned}F_3 &\approx \frac{i}{\hbar}\sum_{\mathbf{q}'}V_{\mathbf{k},\mathbf{k}',\mathbf{q}'}[(N_{\mathbf{k}-\mathbf{q}}+N_{\mathbf{k}'+\mathbf{q}}+1)A_{\mathbf{k},\mathbf{k}',\mathbf{q}-\mathbf{q}'}\\ &\quad-(N_{\mathbf{k}}+N_{\mathbf{k}'}+1)A_{\mathbf{k}-\mathbf{q}',\mathbf{k}'+\mathbf{q}',\mathbf{q}-\mathbf{q}'}]\\ &\quad+\frac{i}{\hbar}\left\{\sum_{\mathbf{k}''\neq\mathbf{k}'+\mathbf{q}}V_{\mathbf{k}'',\mathbf{k}-\mathbf{q},\mathbf{q}}N_{\mathbf{k}}A_{\mathbf{k}'',\mathbf{k}',\mathbf{q}}\right.\\ &\quad+\sum_{\mathbf{k}''\neq\mathbf{k}-\mathbf{q}}V_{\mathbf{k}'+\mathbf{q},\mathbf{k}'',\mathbf{q}}N_{\mathbf{k}'}A_{\mathbf{k},\mathbf{k}'',\mathbf{q}}\\ &\quad-\sum_{\mathbf{k}''\neq\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'',\mathbf{q}}N_{\mathbf{k}-\mathbf{q}}A_{\mathbf{k}''+\mathbf{q},\mathbf{k}',\mathbf{q}}\\ &\quad\left.-\sum_{\mathbf{k}''\neq\mathbf{k}}V_{\mathbf{k}'',\mathbf{k}',\mathbf{q}}N_{\mathbf{k}'+\mathbf{q}}A_{\mathbf{k},\mathbf{k}''+\mathbf{q},\mathbf{q}}\right\}.\end{aligned}\tag{9}$$

It should be noted once more that the procedure of the decoupling used in Eqs. (7)–(9) is not exact. In principle, a more rigorous approach should consist of accounting for all possible Wick contractions. This way a number of the nonclassical correlators of the new types will appear that do not conserve kinetic momentum, such as $\langle a^{\dagger}_{\mathbf{k}}a_{\mathbf{k}'}\rangle, \langle a_{\mathbf{k}}a_{\mathbf{k}'}\rangle, \langle a_{\mathbf{k}}a_{\mathbf{k}'}a_{\mathbf{k}''}a^{\dagger}_{\mathbf{k}'''}\rangle$, etc. These correlators, however, cannot appear spontaneously as a result of the particle-

particle scattering. Their appearance is possible either due to the strong Rayleigh scattering in the system or to the presence of the coherent excitation by two or more laser beams. Both these processes are not considered in the present work.

(4) Finally, the term F_4 involves six creation or annihilation operators corresponding to six different quantum states:

$$F_4 = \frac{i}{\hbar}\Bigg[\sum_{\substack{\mathbf{k}'',\mathbf{q}' \\ \mathbf{k}'',\mathbf{k}-\mathbf{q},\mathbf{k}'+\mathbf{q} \\ \neq \mathbf{k}',\mathbf{k}-\mathbf{q}',\mathbf{k}''+\mathbf{q}'}} V_{\mathbf{k},\mathbf{k}'',\mathbf{q}'} \langle a_{\mathbf{k}''} a^\dagger_{\mathbf{k}-\mathbf{q}'} a^\dagger_{\mathbf{k}''+\mathbf{q}'} a^\dagger_{\mathbf{k}'} a_{\mathbf{k}-\mathbf{q}} a_{\mathbf{k}'+\mathbf{q}} \rangle + \sum_{\substack{\mathbf{k}'',\mathbf{q}' \\ \mathbf{k}'',\mathbf{k}-\mathbf{q},\mathbf{k}'+\mathbf{q} \\ \neq \mathbf{k},\mathbf{k}'-\mathbf{q}',\mathbf{k}''+\mathbf{q}'}} V_{\mathbf{k}',\mathbf{k}'',\mathbf{q}'} \langle a^\dagger_{\mathbf{k}} a_{\mathbf{k}''} a^\dagger_{\mathbf{k}'-\mathbf{q}'} a^\dagger_{\mathbf{k}''+\mathbf{q}'} a_{\mathbf{k}-\mathbf{q}} a_{\mathbf{k}'+\mathbf{q}} \rangle$$
$$- \sum_{\substack{\mathbf{k}'',\mathbf{q}' \\ \mathbf{k}-\mathbf{q}-\mathbf{q}',\mathbf{k}''+\mathbf{q}',\mathbf{k}'+\mathbf{q}' \\ \neq \mathbf{k},\mathbf{k}',\mathbf{k}''}} V_{\mathbf{k}-\mathbf{q},\mathbf{k}'',\mathbf{q}'} \langle a^\dagger_{\mathbf{k}} a^\dagger_{\mathbf{k}'} a^\dagger_{\mathbf{k}''} a_{\mathbf{k}-\mathbf{q}-\mathbf{q}'} a_{\mathbf{k}''+\mathbf{q}'} a_{\mathbf{k}'+\mathbf{q}} \rangle - \sum_{\substack{\mathbf{k}'',\mathbf{q}' \\ \mathbf{k}-\mathbf{q},\mathbf{k}'+\mathbf{q}-\mathbf{q}',\mathbf{k}''+\mathbf{q}' \\ \neq \mathbf{k},\mathbf{k}',\mathbf{k}''}} V_{\mathbf{k}'+\mathbf{q},\mathbf{k}'',\mathbf{q}'} \langle a^\dagger_{\mathbf{k}} a^\dagger_{\mathbf{k}'} a_{\mathbf{k}-\mathbf{q}} a^\dagger_{\mathbf{k}''} a_{\mathbf{k}'+\mathbf{q}-\mathbf{q}'} a_{\mathbf{k}''+\mathbf{q}'} \rangle \Bigg]. \tag{10}$$

We assume that their contribution is negligible ($F_4=0$). Neglecting the terms F_4 means that the phase coherence between six distinct states is negligible because of some finite amount of decoherence in the system. In the case of six-wave mixing experiments, for instance, this approximation will fail. Also, for the polaritons in a random external potential, there are strong high-order correlations between the states with different $\mathbf{k}$, resulting in the inhomogeneous polariton density. This means that the suggested approach cannot describe spatially inhomogeneous systems with induced or spontaneous pattern formation.

The set of Eqs. (5)–(10) describes the dynamics of bosonic systems accounting for particle-particle interactions beyond the Born-Markov approximation. Equations (5)–(10) allow for non-energy-conserving processes and may predict a qualitatively different dynamics of the system with respect to the Boltzmann equations. We remind that the Boltzmann equations only contain the occupation numbers, while in the system (5)–(10), the correlations between different states in the reciprocal space are described by means of the fourth-order nonclassical correlators.

III. DECOHERENCE AND CLASSICAL LIMIT

The formalism derived in the previous section assumes that decoherence is weak enough in order to allow for the conservation of the fourth-order correlator. In this section, we consider the regime of strong decoherence. The decoherence processes are mainly governed by forward scattering of polaritons with acoustic phonons which do not affect directly the occupation numbers. Also, the polariton-polariton forward scattering treated beyond the Born approximation[31] can contribute to the temporal decay of fourth-order correlators. To treat the above mentioned process phenomenologically, one can introduce a decoherence time τ_{dec} in Eq. (6). This approach allows one to describe a smooth transition between the coherent regime described in Sec. II and the Boltzmann limit. It allows the equation for $A_{\mathbf{k},\mathbf{k}',\mathbf{q}}$ to be rewritten in the following form:

$$\frac{dA_{\mathbf{k},\mathbf{k}',\mathbf{q}}}{dt} = \left[\frac{i}{\hbar}(\varepsilon_{\mathbf{k}-\mathbf{q}} + \varepsilon_{\mathbf{k}'+\mathbf{q}} - \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'}) - \frac{1}{\tau_{dec}} \right] A_{\mathbf{k},\mathbf{k}',\mathbf{q}} + F_1 + F_2 + F_3. \tag{11}$$

Though this is not always the case in the experimental situation, let us suppose that the decoherence time is short enough, so that $VN_{tot} \ll 1/\tau_{dec}$, where N_{tot} is the total number of polaritons in the system and V is the mean value of the matrix element. In this case, one can neglect in Eq. (11) the terms corresponding to the energy renormalization F_1 together with the terms F_3. The latter terms can also be neglected because they contain a sum of the fourth-order correlators corresponding to different states in the reciprocal space whose phases are more or less random so that together they yield a zero contribution (random phase approximation). Then, one can assume that due to the strong decoherence the values $A_{\mathbf{k},\mathbf{k}',\mathbf{q}}$ reach their equilibrium much faster than the occupation numbers, i.e., we divide the variables in our system into the slow ones (occupation numbers) and the fast ones (correlators). Such an approach is frequently applied in chemical kinetics, where all the intermediate products of chemical reactions are considered to be in quasiequilibrium (and in our case, the fourth-order correlator is indeed an "intermediate product"). Thus, one can write

$$\frac{dA_{\mathbf{k},\mathbf{k}',\mathbf{k}-\mathbf{q},\mathbf{k}'+\mathbf{q}}}{dt} = \left[\frac{i}{\hbar}(\varepsilon_{\mathbf{k}-\mathbf{q}} + \varepsilon_{\mathbf{k}'+\mathbf{q}} - \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'}) - \frac{1}{\tau_{dec}} \right] A_{\mathbf{k},\mathbf{k}',\mathbf{k}-\mathbf{q},\mathbf{k}'+\mathbf{q}} + F_2 = 0,$$

$$A_{\mathbf{k},\mathbf{k}',\mathbf{k}-\mathbf{q},\mathbf{k}'+\mathbf{q}} = \frac{F_1}{\frac{1}{\tau_{dec}} - \frac{i}{\hbar}(\varepsilon_{\mathbf{k}-\mathbf{q}} + \varepsilon_{\mathbf{k}'+\mathbf{q}} - \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'})}$$
$$= \frac{iV_{\mathbf{k},\mathbf{k}',\mathbf{q}}}{(\varepsilon_{\mathbf{k}-\mathbf{q}} + \varepsilon_{\mathbf{k}'+\mathbf{q}} - \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'}) + i\frac{\hbar}{\tau_{dec}}}[N_{\mathbf{k}}N_{\mathbf{k}'}(N_{\mathbf{k}-\mathbf{q}} + N_{\mathbf{k}'+\mathbf{q}} + 1) - N_{\mathbf{k}-\mathbf{q}}N_{\mathbf{k}'+\mathbf{q}}(N_{\mathbf{k}} + N_{\mathbf{k}'} + 1)]. \tag{12}$$

Substituting this expression for $A_{\mathbf{k},\mathbf{k}',\mathbf{q}}$ into the equation for the occupation numbers [Eq. (5)], one obtains a set of semiclassical Boltzmann equations:

$$\frac{dN_{\mathbf{k}}}{dt} = \sum_{\mathbf{k}',\mathbf{q}} W_{\mathbf{k},\mathbf{k}',\mathbf{q}}[N_{\mathbf{k}-\mathbf{q}}N_{\mathbf{k}'+\mathbf{q}}(N_{\mathbf{k}} + N_{\mathbf{k}'} + 1) - N_{\mathbf{k}}N_{\mathbf{k}'}(N_{\mathbf{k}-\mathbf{q}} + N_{\mathbf{k}'+\mathbf{q}} + 1)], \tag{13}$$

where the scattering rates are given by the following formula:

$$W_{\mathbf{k},\mathbf{k}',\mathbf{q}} = \frac{|V_{\mathbf{k},\mathbf{k}',\mathbf{q}}|^2}{\hbar}\frac{1/\tau_{dec}}{(\varepsilon_{\mathbf{k}-\mathbf{q}} + \varepsilon_{\mathbf{k}'+\mathbf{q}} - \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'})^2/\hbar^2 + 1/\tau_{dec}^2}. \tag{14}$$

One can see from Eq. (14) that the scattering rate is the fastest for the energy-conserving processes, where it is simply given by the Fermi golden rule, $W_{\mathbf{k},\mathbf{k}',\mathbf{q}} = |V_{\mathbf{k},\mathbf{k}',\mathbf{q}}|^2\tau_{dec}/\hbar$. For the non-energy-conserving processes, the probability of scattering is reduced by a standard Lorentzian factor.

IV. PARTICULAR CASE OF A POLARITON PARAMETRIC OSCILLATOR

A. Formalism and parameters

The complete set of kinetic Eqs. (5)–(10) is extremely complicated and, for the general case of nonresonant pumping, requires hard numerical modeling. In the present section, we consider the simple example of polariton parametric amplifier (PPO) involving only three quantum states. Due to the strong nonparabolicity of the lower polariton branch, a pair of exciton-polaritons created by the pump pulse at the so-called magic angle scatters into nondegenerate signal and idler states with both energy and momentum conserved as is shown in Fig. 1. Our goal is to describe the transition from the Boltzmann limit to the coherent regime. We do not consider here in detail the selection of the signal-idler states among the continuum of polariton states. The selection problem, however, is far from being trivial because of strong renormalization of the polariton branches with the increase of the pump intensity. At high polariton density, the conservation of energy and momentum takes place for the states essentially different from those in the low density limit.[22,23] Bearing this in mind, we consider here the simplest model based on the assumption that the PPO may be described by only three quantum states, namely, the pump, signal, and idler states. An advantage of our formalism with respect to the precedent ones derived in the same spirit[14,24] is that it allows to take into account simultaneously signal-idler correlation and pump depletion. The Hamiltonian of this system can be written in the following form:

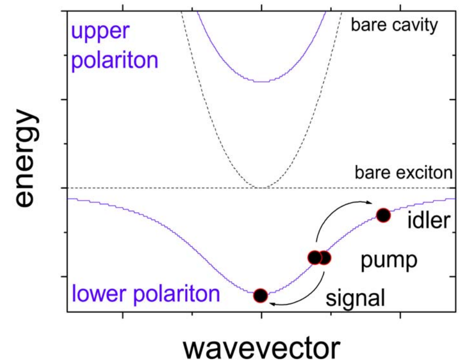

FIG. 1. (Color online) Polariton dispersion (blue) and schematic parametric scattering of two pump polaritons into signal and idler polaritons at the "magic angle." The bare cavity and exciton energies are shown dashed.

$$H = (\varepsilon_s a_s^\dagger a_s + \varepsilon_i a_p^\dagger a_p + \varepsilon_i a_i^\dagger a_i) + U(|X_s|^2 a_s^\dagger a_s + |X_p|^2 a_p^\dagger a_p + |X_i|^2 a_i^\dagger a_i)(|X_s|^2 a_s^\dagger a_s + |X_p|^2 a_p^\dagger a_p + |X_i|^2 a_i^\dagger a_i) + (Va_p^\dagger a_p^\dagger a_s a_i + V^* a_p a_p a_s^\dagger a_i^\dagger), \tag{15}$$

where the indices p, s, and i correspond to the pump, signal, and idler, respectively. The first term describes free particles, the second one describes energy blueshifts (X is the Hopfield coefficient corresponding to the percentage of the exciton fraction in each of the states), and the third term corresponds to the parametric process. Equations (5)–(9) reduce in this case to

$$\frac{dN_s}{dt} = -\frac{N_s}{\tau_s} + \frac{2}{\hbar}\,\mathrm{Im}\{V\langle a_s^\dagger a_i^\dagger a_p a_p\rangle\} = -\frac{N_s}{\tau_s} + \frac{2}{\hbar}\,\mathrm{Im}\{VA\}, \tag{16a}$$

$$\frac{dN_i}{dt} = -\frac{N_i}{\tau_i} + \frac{2}{\hbar}\,\mathrm{Im}\{VA\}, \tag{16b}$$

$$\frac{dN_p}{dt} = -\frac{N_p}{\tau_p} - \frac{4}{\hbar}\,\mathrm{Im}\{VA\} + P\frac{\hbar^2\tau_p^{-2}}{\delta_P^2 + \hbar^2\tau_p^{-2}}, \tag{16c}$$

$$\frac{dA}{dt} = -\left(\frac{1}{2\tau_s} + \frac{1}{2\tau_i} + \frac{1}{\tau_p} + \frac{1}{\tau_{dec}}\right)A + \frac{i}{\hbar}\delta_{PPO}A + \frac{iV}{\hbar}[N_p^2(N_s + N_i + 1) - 4N_sN_i(N_p + 1)], \tag{16d}$$

where we have introduced the polariton lifetimes and external pumping term P; E_0 is the energy of the pump laser and τ_{dec} is the decoherence time. If pump depletion and decoherence are neglected, we recover the results obtained earlier by Ciuti *et al.*[14]

One can see that there are two energy detunings that play a crucial role. The first one, in Eq. (16c), is the energy dif-

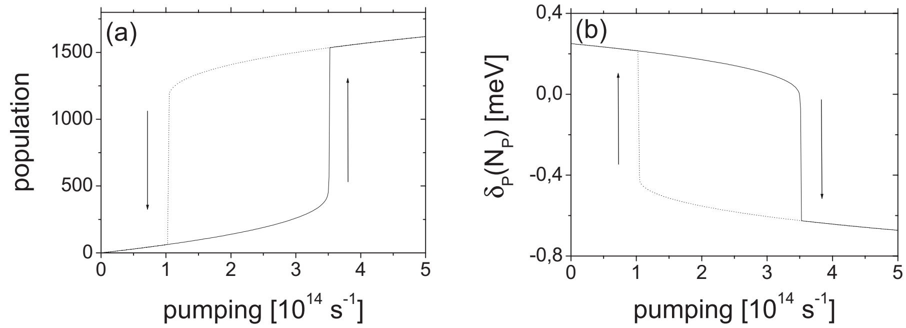

FIG. 2. Single mode model for a positive detuning $\delta_p(0)=0.25$ meV. (a) Pump population versus pump intensity which is adiabatically increased (solid line) or decreased (dashed line). (b) Detuning $\delta_p(N_p)$ versus the pump intensity adiabatically increased (solid line) and decreased (dashed line).

ference between the laser and the renormalized pumped polariton state. It is responsible for the bistable behavior of the PPO and will be referred to as the pump detuning in our further consideration,

$$\delta_p(N_p) = E_0 - \varepsilon_p - U|X_p|^2(|X_p|^2 N_p + |X_s|^2 N_s + |X_i|^2 N_i). \tag{17}$$

The second detuning, in Eq. (16d), represents the dynamical energy mismatch between renormalized signal, pump, and idler states of the PPO:

$$\delta_{PPO}(N_i,N_s,N_p) = (\varepsilon_s + \varepsilon_i - 2\varepsilon_p) + U(|X_i|^2 + |X_s|^2 - 2|X_p|^2) \times(|X_i|^2 N_i + |X_s|^2 N_s + |X_p|^2 N_p). \tag{18}$$

In our numerical simulations, we consider a realistic case of a GaAs microcavity similar to that in Ref. 11. The Rabi splitting is 6 meV and the cavity photon lifetime is $\tau_{ph}=2$ ps. The signal, pump, and idler lifetimes are respectively given by

$$\tau_{s,p,i} = \frac{\tau_{ph}}{(1-|X_{s,p,i}|^2)}. \tag{19}$$

The exciton-exciton matrix element of interaction is taken as[25] $U=6E_b(a_b^2/S)$, where $E_b=10$ meV is the exciton binding energy, $a_b=100$ Å the two-dimensional (2D) exciton Bohr radius, and S the surface of the laser spot for which we take a lateral size of 5 μm.

B. Single mode dynamics under the cw laser pumping

In order to separate the contributions of the two main nonlinearities in the system, we first consider the case of a single mode system with $\delta_p(0)>0$. Figure 2(a) shows the steady state polariton population versus cw pumping whose energy lies above the bare energy of the pumped state. cw pumping is adiabatically turned on (solid line) and turned off (dashed line). Figure 2(b) shows $\delta_p(N_p)$ for the same pumping conditions. One can see the typical hysteresis cycle of a bistable system. At low pumping, N_p depends superlinearly on P. The absorption of light by the mode decreases the value of $|\delta_p(N_p)|$, which, in turn, results in the increase of the absorption. This process finds its paroxysm at the turning point of the curve where $\delta_p(N_p)$ changes sign. In the model case which we consider here, the pump population jumps by a factor of 5 at this point. Above this point, N_p depends sublinearly on P since further increase of pumping provokes an increase of $|\delta_p(N_p)|$, which limits the increase of absorption. If P is decreasing, similar processes take place except that the turning point is situated at lower P than in the case of increasing pumping. All these processes result in the typical hysteresis cycle shown in Fig. 2. The hysteresis strongly affects the dynamics of the PPO as has been first outlined in Refs. 22 and 23, and as we shall see in the next sections.

C. Dynamics of three modes under steplike onset at $t=0$

We first focus on the establishment of the steady state regime under cw pumping switching at $t=0$. Figure 3(a) shows the signal and the pump state populations versus the pump laser intensity. The pump laser energy is now taken to be resonant with the bare polariton energy so that no bistability is expected. We also take $\delta_{PPO}(0,0,0)=0$. The parametric process starts to be efficient for rather low pump intensities and small polariton populations, so that the corresponding value of δ_p remains smaller than the linewidth [Fig. 3(b)]. Figure 3(c) shows the temporal dynamics of the signal and pump states under cw excitation far above threshold. The buildup time of the signal is about 50 ps and it is longer than that of the pump state. However, the signal dynamics is quite smooth and shows no oscillations. In this regime, we do not expect that the Boltzmann approach would give qualitatively different results, except for the weak maxima at about 25 ps for the pump and at 50 ps for the signal. One can observe in Fig. 3(a) that N_p continues to grow even above the nonlinear threshold. This contrasts with the results of the previous models, which neglect the pump dynamics and fix the pump population above threshold.

The same quantities as in Fig. 3 are shown in Figs. 4(a)–4(d) for the pump detunings $\delta_p(0)=1$ and 0.25 meV. In Fig. 4(a), the pump and signal populations show a nonlinear dependence on the laser intensity. Two very different regimes can be achieved depending on the value of the pump detuning. If the detuning is large, the PPO threshold is reached before the bistability threshold (solid lines). In this case, there are two jumps in the signal intensity as a function of pumping intensity, as is shown by the black curve. On the contrary, for smaller values of $\delta_p(0)$, the bistable threshold takes place before the PPO threshold (dashed lines). In that case, only one intensity jump is observed for both pump and signal intensities. However, this result and the next one can be altered if we would consider a realistic 2D microcavity.[24]

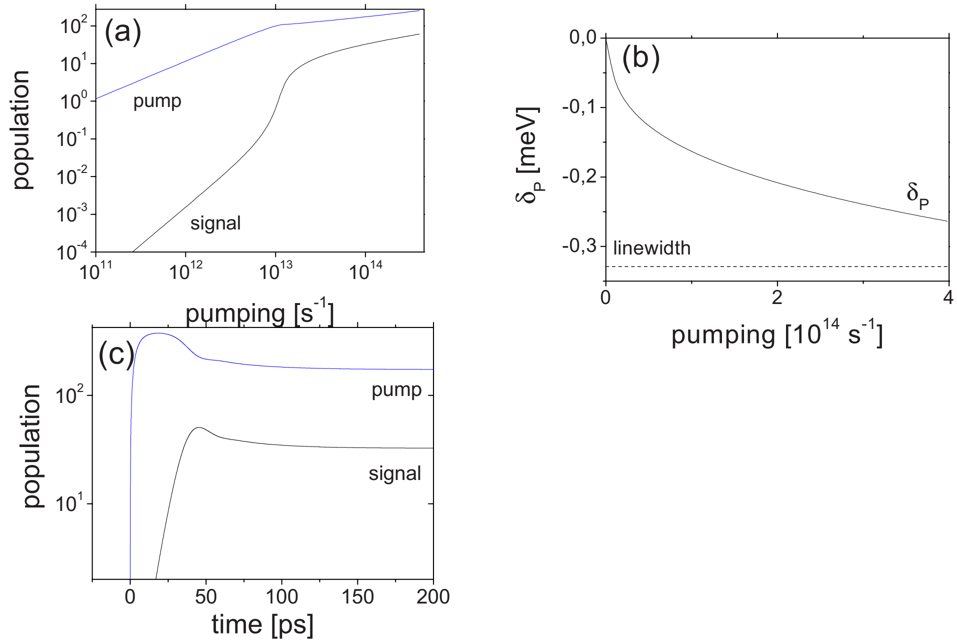

FIG. 3. (Color online) Three modes model with resonant excitation of the pump state [$\delta_p(0)=0$]. (a) Steady state signal (black) and pump populations (blue) versus the cw pump intensity which is adiabatically enhanced. (b) Detuning $\delta_p(N_p)$ versus the pump intensity adiabatically enhanced. (c) Time dependence of the signal (black) and pump populations (blue) for a pumping intensity of 10^{14} particles/s. The corresponding particle density in the steady state is 2.3×10^9 cm^{-2}.

The time domain results are shown in Fig. 4(c) for $\delta_p(0)=1$ meV and in Fig. 4(d) for $\delta_p(0)=0.25$ meV. The signal and pump intensities show abrupt jumps once the threshold is reached for both detunings. The populations show oscillations after reaching the threshold, demonstrating the important role played by the correlations between signal, pump, and idler states in this regime. These oscillations are damped because of the continuous filling of the pump state by the external laser beam. For moderate pumping and at large detuning, two successive thresholds can be seen in Fig. 4(c) (solid lines) for the signal and pump populations. The dotted lines show the population numbers for pumping below threshold. In the case of high laser intensities, these two thresholds take place simultaneously (not shown). In Fig. 4(d), the PPO starts after passing the bistable threshold and leads to a jump of the signal intensity. For pumping below the threshold, the signal state is filled only by spontaneous scattering and remains weak for the two detunings.

D. Kick effect

In this section, we consider the effect of a short kick pulse resonant with the pump mode which comes after the establishment of the steady state. We consider the case $\delta_p(0)=1$ meV. The intensity of the cw pumping laser is chosen in order to maintain the system just below the bistable thresh-

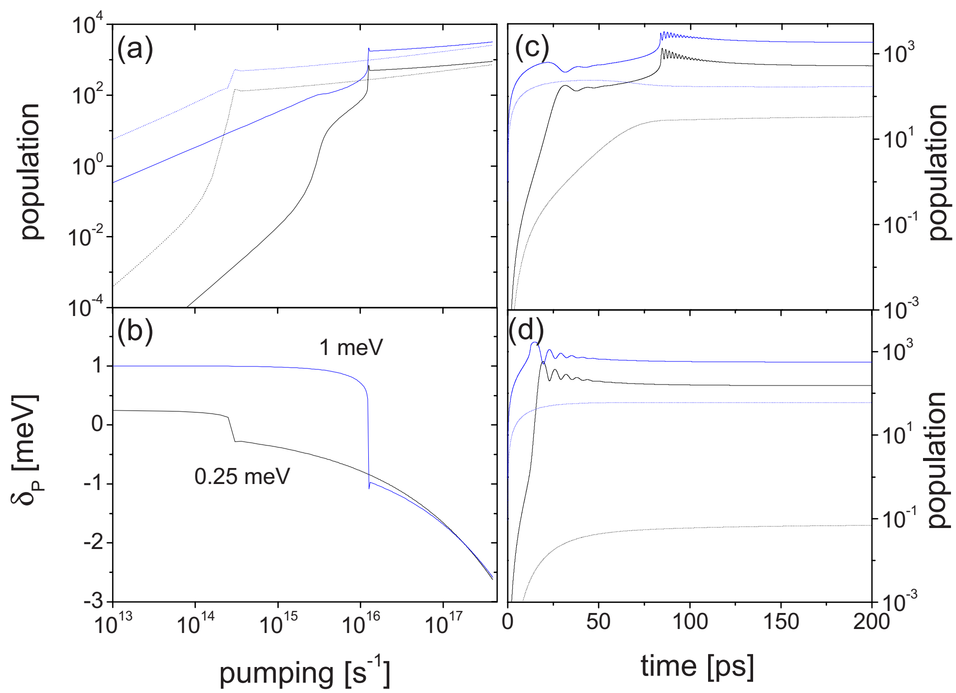

FIG. 4. (Color online) Three modes model for the positive pump detuning $\delta_p(0)$. (a) Steady state signal (black) and pump populations (blue) for $\delta_p(0)=0.25$ meV (dashed) and $\delta_p(0)=1$ meV (solid). The cw pump intensity shown on the y axis is adiabatically increased. (b) Detuning $\delta_p(N_p)$ versus the pump intensity for $\delta_p(0)=0.25$ meV (black) and $\delta_p(0)=1$ meV (blue). (c) $\delta_p(0)=1$ meV. Time dependence of the signal (black) and pump populations (blue) for a pumping intensity of 2×10^{16} particles/s (dashed) and 7×10^{16} particles/s (solid). (d) $\delta_p(0)=0.25$ meV. Time dependence of the signal (black) and pump populations (blue) for a pumping intensity of 10^{14} particles/s (dashed) and 10^{15} particles/s (solid).

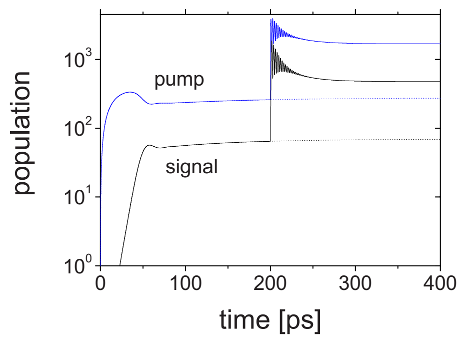

FIG. 5. (Color online) Kick effect for the three modes model. Time dependence of signal (black) and pump state populations (blue). The pumping is composed of a cw laser [$\delta_p(0)=1$ meV, 1×10^{16} particles s^{-1}] and a short kick pulse at $t=100$ ps (2.5×10^{17} particles s^{-1}). The dashed lines correspond to the populations without kick.

old. The intensity emitted by the signal is, therefore, rather weak. Then a 1 ps long kick pulse is sent to the pump state. It induces the increase of the pump intensity sufficient to pass the bistable threshold, as Fig. 5 shows. Consequently, after the arrival of the kick, the signal intensity increases by almost 1 order of magnitude. Remarkably, the system does not go back to its initial state after the kick pulse has passed, but it stabilizes to a new equilibrium state characterized by an intense signal emission. This effect can be used for the realization of low threshold optical switches.

E. From quantum to classical limit

Figure 6 shows the impact of the decoherence on the dynamics of the PPO. It can be clearly seen that the decrease of the decoherence time leads to the increase of both bistable and PPO thresholds. In the time domain, it leads to the washing out of the oscillations in pump and signal intensities due to the suppression of the nonclassical correlations between them. It is also seen that the decoherence makes longer the time needed for the pump and signal states to reach stationary values under cw excitation. It is clearly seen from Fig. 6(c) that in the limit of the small decoherence times, the system recovers the Boltzmann-limit dynamics.

V. CONCLUSIONS

In conclusion, we have derived a closed set of kinetic equations describing a system of interacting bosons beyond the Markovian approximation. The dynamics of the occupation numbers is shown to be strongly altered by the buildup of the nonclassical four-particle correlators. The decoherence process leading to the fast suppression of these correlators is shown to provoke the transition from the quantum to the classical limit. This system of kinetic equations is applied to the dynamics of a three state polariton parametric oscillator. The general equation set which is obtained by us is quite heavy, and its solution requires great numerical effort. However, this formalism has an important advantage of taking into account the incoherent phonon dynamics and the coherent nature of the polariton-polariton scattering process in the presence of macroscopically occupied polariton modes. It represents a bridge between the fully coherent picture (Gross-Pitaevskii equations) and the fully incoherent picture (Boltzmann approach).

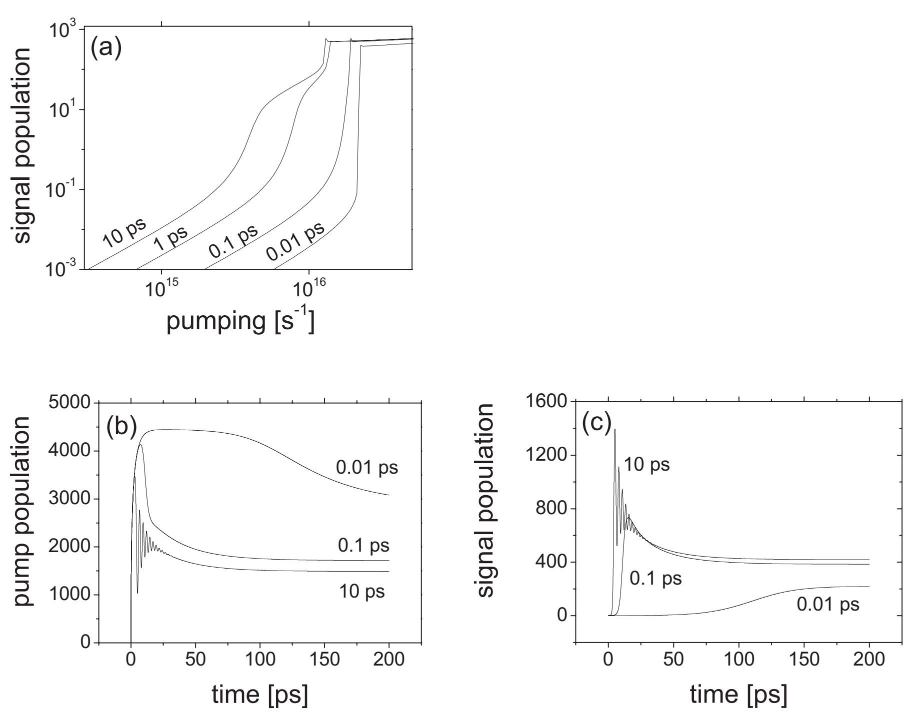

FIG. 6. Signal and pump populations versus the pump intensity for different decoherence times [$\delta_p(0)=1$ meV]. The condition $VN_{tot}\ll\hbar/\tau_{dec}$ is satisfied for decoherence times shorter than 0.1 ps. (a) Steady state population of the signal state. (b) Time dependence of the signal population (2.5×10^{16} particles s^{-1}). (c) Time dependence of the pump population (2.5×10^{16} particles s^{-1}).

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