Dynamical Theory of Polariton Amplifiers

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We present the theory of the dynamics of the polariton amplifier in the region of small polariton densities. We give an analytical solution for the polariton condensate density matrix and show that the formation of a coherent quantum state is possible. Once the condensate is formed, the coherence becomes macroscopically long living. Polariton amplifier represents, therefore, an optical memory element, where the input weak coherent signal can be amplified and kept.

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Introduction.—Conventional lasers need an inversion of population in the electronic system and are characterized by a threshold density of emission above which the stimulated radiation dominates over absorption. The lasing optical mode above the threshold is described as a nonlinear oscillator, which is resonantly excited and stabilized due to nonlinearity, and the laser light is produced in a randomly phased coherent state [1]. The formation of a coherent state is the key feature of the laser emission, as compared to the other monochromatic light sources.

A device usually referred to as a polariton laser [2] consists of a semiconductor microcavity maintaining the strong coupling regime, so that two exciton-polariton dispersion branches exhibit avoided crossing [3]. Exciton-polaritons excited by electric or nonresonant optical pumping relax along the dispersion curves down to the ground state, which is the lower polariton branch state characterized by the in-plane momentum $\mathbf{k} = 0$ (see Fig. 1). This relaxation can be stimulated if the ground-state population exceeds 1. Such stimulation is a consequence of the bosonic nature of polaritons, as evidenced in a number of experiments [4–6]. Bosonic stimulation of polariton relaxation does not *a priori* imply the Bose-Einstein condensation (BEC) in the final state, but it does not exclude it either. In spite of the frequent use of terms "polariton laser," "polariton amplifier," and "polariton condensation" in recent publications, there is still uncertainty about their exact meaning. That is why we find it important to clearly formulate what we mean by using these terms.

In what follows we refer to the devices based on the bosonic amplification of polariton relaxation towards the ground state as polariton amplifiers. If such amplification appears spontaneously and leads to a macroscopic population of the ground state, it will be called the polariton lasing. Because of a very small polariton effective mass, $m^* \sim 10^{-4} m_0$, the characteristic temperature T_0 for polariton lasing is high. For a large system area S, with the logarithmic precision one has [7] $T_0 \sim 2\pi\hbar^2 n / k_B m^* \ln(nS)$, where n is the polariton concentration, and the polariton lasing is expected to happen up to the room temperature [8]. Finally, if a coherent state is formed by the polaritons at $\mathbf{k} = 0$, this will be referred to as condensation. The formation of the polariton condensate reveals itself in the higher-order correlation functions of the emitted light. In particular, it can be seen from the intensity autocorrelation observed in the Hanbury Brown and Twiss experiment [9]. This autocorrelation function was measured recently for the polariton laser by Deng *et al.* [10].

The effect of spontaneous formation of a polariton condensate must be essentially nonlinear as only interactions between polaritons (direct or indirect) may allow them to change their statistical distribution. Thus, this effect is excluded at low concentrations of exciton-polaritons (further we will give the criterion for "small" concentration). On the other hand, if an initial seed of coherent polaritons is introduced by a resonant optical excitation into the ground state, it may induce formation of a polariton condensate, stimulating the relaxation of excited polaritons to the state already characterized by a given coherence. This is an effect of linear amplification that may take place in any polariton amplifier and that does not imply any polariton-polariton interaction in the condensate. As we show in this Letter, neglecting polariton-polariton interaction allows one to describe this effect within a simple model of a damped quantum

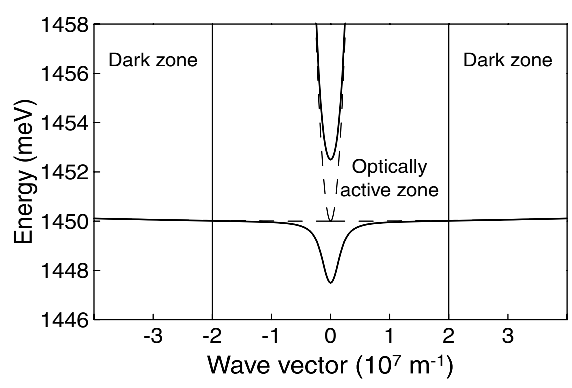

FIG. 1. The dispersion of uncoupled photons and excitons (dashed lines) and microcavity polaritons in the strong-coupling regime (solid lines) in a typical GaAs-based cavity.

oscillator, so that time-dependent polariton statistics can be obtained analytically.

It should be noted that the regime of low polariton density is a peculiarity for polariton amplifiers with respect to conventional lasers. Polariton amplifiers allow one to work with small quantities of photons revealing quantum characteristics of emitted light. Nonlinear effects in polariton lasers pointed out in the literature, e.g., the renormalization of the polariton dispersion [11,12] and self-interaction within the quasicondensate [13], are not important in the low-density regime we consider.

We show below that the possibility of the polariton condensate formation has a kinetic origin, and the survival of the initial coherent seed is related to a remarkable *dynamic* instability inherent to the polariton condensate formation. This instability allows the amplification of the ground-state population preserving the coherence of the condensate. Moreover, if the relaxation speed is high enough, so that the instability is realized, the coherence of the condensate remains macroscopically long living in the steady-state regime, that makes polariton amplifiers extremely promising for applications as quantum memory elements. Since the possibility of BEC in the polariton amplifier has a dynamical origin, the theory formulated below describes the quantum kinetic of polariton relaxation and the coherent state buildup.

Polariton amplifier dynamics.—We consider the polaritons as perfect bosons. The pseudospin of polaritons (or the related light polarization) is given by the pumping light and is conserved during the polariton relaxation on the time scale of interest [14]. In the case of noninteracting polaritons, the kinetic equation for the condensate density matrix ρ_0 has the same structure as the equation proposed by Landau [15] for the harmonic oscillator. In the interaction representation it reads

$$\dot{\rho}_0 = -\tfrac{1}{2}[W_{\rm out}(t)(a_0^\dagger a_0\rho_0 + \rho_0 a_0^\dagger a_0 - 2a_0\rho_0 a_0^\dagger) + W_{\rm in}(t)(a_0 a_0^\dagger\rho_0 + \rho_0 a_0 a_0^\dagger - 2a_0^\dagger\rho_0 a_0)]. \quad (1)$$

Here $a_0^\dagger$ and a_0 are the creation and annihilation operators for polaritons in the condensate. The BEC kinetics is described by Eq. (1) on a time scale much greater than the typical duration of the relaxation events, which change the occupation of the condensate by ± 1, and these events are assumed to be not correlated (the Markov approximation). The coefficients $W_{\rm in}(t)$ and $W_{\rm out}(t)$ define the income and outcome rates for polaritons in the condensate, as it is seen from the equation for the average number of the condensed polaritons $N_0 = \langle a_0^\dagger a_0\rangle$. According to Eq. (1) one has $\dot{N}_0 = W_{\rm in}(t)(N_0 + 1) - W_{\rm out}(t)N_0$.

There are important differences between Eq. (1) and those used for the lasing mode [1,16–18]. In the case of polariton relaxation, the rates $W_{\rm in}(t)$ and $W_{\rm out}(t)$ depend on time through the occupation numbers $N_{\mathbf{k}}(t)$ of the polaritons which are not in the condensate ($\mathbf{k} \neq 0$). The account for this time dependence makes the condensate dynamics nonlinear and it is necessary to describe the above mentioned dynamic instability. On the other hand, in contrast to the conventional laser [1], we do not allow for the fourth power in a_0 and $a_0^\dagger$ terms, describing the interactions within the condensate. Therefore, our theory applies to the region of low occupation numbers N_0. More precisely, we consider the dynamics of the condensate in the region $V_0N_0/\hbar \ll W_{\rm in,out} \sim \Gamma_0$, where the typical values of the rates are given by the lifetime of a polariton in the condensate Γ_0^{-1}, and V_0 is the interaction energy of polaritons at $\mathbf{k} = 0$ [12]. We show below that it is the low occupation regime where the coherence of the condensate is built up, which allows one to study this phenomenon within the framework of Eq. (1).

The relaxation of polaritons can be accompanied by the appearance of "the order parameter" $\alpha(t) = \langle a_0\rangle$, which defines, as we see below, the coherent properties of the condensate. To study the possibility of the order parameter formation, we introduce a "seed"; i.e., we look for the solution of Eq. (1) at the initial condition $\rho_0(0) = |\alpha_0\rangle\langle\alpha_0|$. The initial coherent state of the condensate is defined as usual [19] by the action of the unitary shift operator $D(\alpha_0) = \exp\{\alpha_0 a_0^\dagger - \alpha_0^* a_0\}$ on the vacuum state, $|\alpha_0\rangle = D(\alpha_0)|0\rangle$.

A direct method to solve Eq. (1) is based on the introduction of the ordering index for operators and then applying Feynman's disentangling procedure [20]. The goal, however, can be reached more easily by the observation that if one solution, $\varrho_0(t)$, of Eq. (1) is known, one can construct other solutions (for different initial conditions) as $D(\alpha)\varrho_0 D(\alpha)^\dagger$, provided that $\dot{\alpha} = (1/2)(W_{\rm in} - W_{\rm out})\alpha$. Therefore, the problem is reduced to finding the solution ϱ_0 with $\varrho_0(0) = |0\rangle\langle 0|$. This particular solution can be written as $\varrho_0(t) = f(a_0^\dagger a_0, t)$, and it follows from Eq. (1) and the relation $a_0 f(a_0^\dagger a_0, t) = f(a_0^\dagger a_0 + 1, t)a_0$ that $f(z, t)$ satisfies the differential-difference equation

$$\dot{f} = W_{\rm out}(t)[(z+1)f(z+1,t) - zf(z,t)] + W_{\rm in}(t)[zf(z-1,t) - (z+1)f(z,t)], \quad (2)$$

which can be solved by standard methods. In particular, the inverse Laplace transformation with respect to variable z (or the generating function method of Ref. [17]) turns Eq. (2) into the equation in the first-order partial derivatives, solvable with the method of characteristics. The final answer is

$$\rho_0 = \frac{1}{1+c} D(\alpha)\exp\left\{-\ln\left[\frac{1+c}{c}\right]a_0^\dagger a_0\right\}D(\alpha)^\dagger, \quad (3)$$

where the temporal dependencies are given by

$$\alpha(t) = \alpha_0 e^{g(t)}, \qquad c(t) = e^{2g(t)}\int_0^t W_{\rm in}(\tau)e^{-2g(\tau)}d\tau, \quad (4)$$

$$g(t) = \frac{1}{2}\int_0^t [W_{\rm in}(\tau) - W_{\rm out}(\tau)]d\tau. \quad (5)$$

In a particular case of a 2D system of noninteracting bosons relaxing by acoustic phonon emission, our solution for the order parameter reduces to that derived by Bányai and Gartner [21].

The above solution can be used to calculate the second-order correlator $\langle a_0^\dagger a_0^\dagger a_0 a_0 \rangle$, which can be measured in the two-photon coincidence counting experiments [9,10]. It turns out that this correlator is completely defined by the order parameter $\alpha(t)$. It is convenient to introduce the parameter

$$\eta(t) = \frac{2N_0^2 - \langle a_0^\dagger a_0^\dagger a_0 a_0 \rangle}{N_0^2} \equiv \frac{|\alpha(t)|^4}{N_0^2}, \tag{6}$$

which provides information about the coherence of ground-state polaritons [22], and will be referred to shortly as the coherence degree. This parameter is equal to 1 for the polaritons being in a coherent state, while it goes to 0 for a thermal quantum state [19].

Before presenting the numerical results, we give a qualitative analysis of the polariton condensate formation. First we note that without an initial seed ($\alpha_0 = 0$), the ground-state polaritons always form a thermal quantum state with $\alpha(t) = \eta(t) = 0$. The kinetics of polariton relaxation is characterized by a transient regime, during which the polaritons come to the ground state, after being excited in some $\mathbf{k} \neq 0$ state at $t = 0$. Their relaxation speed depends nonlinearly on the pumping intensity. For a strong enough pumping, the stimulated scattering of polaritons into the ground state flares up at a time $t_s > 0$, so that the income rate increases drastically and becomes much greater than the outcome rate. In the time domain where $W_{\mathrm{in}}(t) > W_{\mathrm{out}}(t)$, the $\alpha = 0$ solution becomes unstable. In the case of a small seed $\alpha_0 \neq 0$ introduced initially, this instability allows the condensation to happen. The initial order parameter survives and is amplified, so that in the steady-state regime one has a large α, large occupation $N_0 = c + |\alpha|^2$, and a finite coherence parameter η.

After the steady-state regime is reached, the $\alpha = 0$ point becomes stable again, since the rates achieve the time independent values $W_{\mathrm{in}}^{\mathrm{st}}$ and $W_{\mathrm{out}}^{\mathrm{st}}$ with $W_{\mathrm{in}}^{\mathrm{st}} < W_{\mathrm{out}}^{\mathrm{st}}$. However, the difference of the stationary rates is very small, inversely proportional to the system area S, which corresponds to a large ground-state population $N_0^{\mathrm{st}} = W_{\mathrm{in}}^{\mathrm{st}}/(W_{\mathrm{out}}^{\mathrm{st}} - W_{\mathrm{in}}^{\mathrm{st}}) \propto S$. If the coherence is formed, its decrease is extremely slow in large cavities.

Numerical results.—The coefficients $W_{\mathrm{in}}(t)$ and $W_{\mathrm{out}}(t)$ for our system are given by [23]

$$W_{\mathrm{in}}(t) = \sum_{\mathbf{k}\neq 0} w_{\mathbf{k}\to 0} N_{\mathbf{k}}(t), \tag{7}$$

$$W_{\mathrm{out}}(t) = \Gamma_0 + \sum_{\mathbf{k}\neq 0} w_{0\to\mathbf{k}} [N_{\mathbf{k}}(t) + 1]. \tag{8}$$

Here $w_{\mathbf{k}'\to\mathbf{k}}$ describes the rate of polariton transitions between the $\mathbf{k}'$ and $\mathbf{k}$ states. Details of the calculation of these scattering rates are given in Ref. [24]. The polariton populations $N_{\mathbf{k}}(t)$ are found from the semiclassical kinetic equation [23,24].

We consider a GaAs-based microcavity containing a single quantum well. The parameters are the same as in Ref. [23]. We assume that this structure contains an equilibrium free electron gas of density 10^{10} cm^{-2}, which speeds up the relaxation processes [23]. The cavity has a finite lateral radius $R = 100$ μm. We model the following experiment: at $t = 0$ an ultrashort laser pulse generates a coherent ground state containing a variable polariton number (seed). At the same time, an incoherent nonresonant cw pumping is turned on. Three pumping densities (0.8, 8, and 160 W/cm^2) are considered. In all cases the strong coupling regime is maintained. Figure 2 shows the evolution of the ground-state population for different pumping densities and a seed of 100. Table I reports the parameters obtained in the steady-state regime (achieved about 1 ns after the pumping start): the ground-state population, the ratio of populations of the ground state and the first excited state (with the wave vector $k = \pi/R$), the ratio of the ground-state population and the total population, and the chemical potential $\mu = k_B T \ln(1 - 1/N_0)$. The temperature T of the polariton subsystem in the steady state becomes very close to the lattice temperature.

It is seen from Fig. 2 that at low pumping density, the seed disappears on a time scale of a few tens of picoseconds. In the intermediate and high pumping densities, the coherence survives, and the buildup of the order parameter accompanies the amplification of the initial

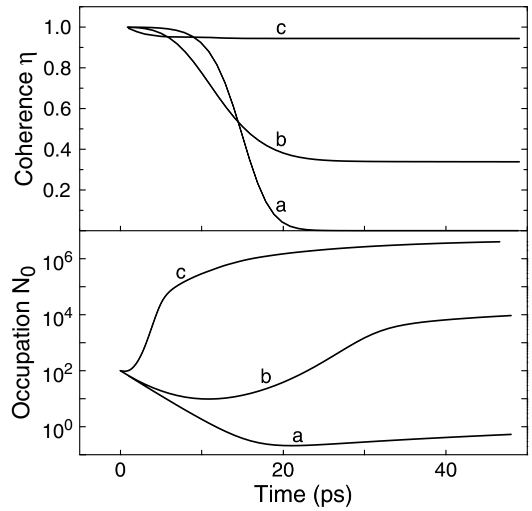

FIG. 2. The evolution of the ground state population N_0 and the coherence parameter η at the initial stage of the condensate formation. The pumping densities are 0.8, 8, and 160 W/cm^2 for curves a, b, and c, respectively.

TABLE I. Main steady-state values obtained numerically.

Pumping density (W/cm^2)	N_0	N_0/N_1	N_0/N	$\lvert\mu\rvert$ (μeV)
0.8	7.7×10^3	23.5	0.04	56
8	2.7×10^5	510	0.59	1.6
160	5.8×10^6	5800	0.95	0.07

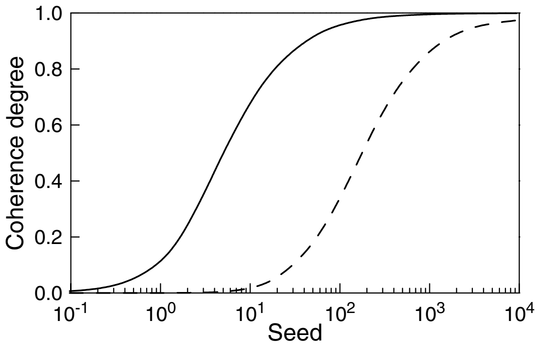

FIG. 3. The ground state coherence degree versus the seed population in the steady-state regime. Nonresonant pumping densities are 8 W/cm^2 (dashed line) and 160 W/cm^2 (solid line).

seed coherent state. This is to our knowledge the first theoretical description of a polariton coherence buildup in a realistic semiconductor microcavity, i.e., including real polariton dispersions and all the major interactions. This coherence buildup is a clear signature of polariton condensation.

In the intermediate pumping densities, the values of the steady-state coherence degree depend noticeably on the seed characteristics. The dependencies of the coherence degree η on the seed population are shown in Fig. 3 for two pumping densities. One can see that the buildup of coherence takes place only if the seed population exceeds some critical value which depends on pumping, but remains always very small (from 10 to 100 polaritons as compared to $N_0 \sim 10^6$ in the steady state).

The polariton condensate is formed and is stabilized at a very early stage of the relaxation processes, where the occupation of the ground state is still small. This allows us to neglect the interaction of polaritons in the condensate to study the coherence evolution. Clearly, at high pumping densities the ground state occupation becomes high, and the interaction of polaritons cannot be omitted. The effect of interaction, however, does not change qualitatively the above picture. The dynamics of the condensate formation will still be characterized by a short, transient time domain, where the coherence is built up (and this domain is adequately described by the above theory), and a macroscopically long time domain, where this coherence and the order parameter relax. The repulsive polariton-polariton interaction will be responsible for the formation of the superfluid phase through the Kosterlitz-Thouless transition. And, if the temperature is less than the transition temperature, the relaxation of the order parameter will remain very slow for large cavities.

In conclusion, we have presented a quantum kinetic theory describing the dynamical aspects of the polariton amplifier in a nonresonantly pumped cavity. A fast relaxation kinetics of the polaritons allows the formation of a coherent quantum state. The coherence is then maintained over macroscopically long time in the steady-state regime.

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[1] L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics* (Cambridge University, Cambridge, England, 1995).
[2] A. Imamoḡlu *et al.*, Phys. Rev. A **53**, 4250 (1996).
[3] For a review, see G. Khitrova *et al.*, Rev. Mod. Phys. **71**, 1591 (1999).
[4] P. G. Savvidis *et al.*, Phys. Rev. Lett. **84**, 1547 (2000); P. G. Lagoudakis *et al.*, Phys. Rev. B **65**, 161310 (2002).
[5] R. M. Stevenson *et al.*, Phys. Rev. Lett. **85**, 3680 (2000).
[6] M. Saba *et al.*, Nature (London) **414**, 731 (2001).
[7] W. Ketterle and N. J. van Druten, Phys. Rev. A **54**, 656 (1996).
[8] G. Malpuech *et al.*, Appl. Phys. Lett. **81**, 412 (2002); M. Zamfirescu *et al.*, Phys. Rev. B **65**, 161205 (2002).
[9] R. Hanbury Brown and R. Q. Twiss, Nature (London) **177**, 27 (1956); **178**, 1046 (1956).
[10] H. Deng *et al.*, Science **298**, 199 (2002).
[11] I. V. Beloussov and V. V. Frolov, Phys. Rev. B **54**, 2523 (1996); C. Ciuti *et al.*, Phys. Rev. B **62**, R4825 (2000).
[12] D. Porras and C. Tejedor, Phys. Rev. B **67**, 161310 (2003).
[13] M. Holland *et al.*, Phys. Rev. A **54**, R1757 (1996); C. W. Gardiner and P. Zoller,*ibid.* **58**, 536 (1998); F. Tassone and Y. Yamamoto,*ibid.* **62**, 063809 (2000).
[14] M. D. Martín *et al.*, Phys. Rev. Lett. **89**, 077402 (2002).
[15] L. D. Landau, Z. Phys. **45**, 430 (1927).
[16] Y. R. Shen, Phys. Rev. **155**, 921 (1967).
[17] B. Ya. Zel'dovich, A. M. Perelomov, and V. S. Popov, Zh. Eksp. Teor. Fiz. **55**, 589 (1968) .[Sov. Phys. JETP **28**, 308 (1969)].
[18] A. Imamoḡlu and R. J. Ram, Phys. Lett. A **214**, 193 (1996).
[19] R. Glauber, *Quantum Optics and Electronics*, edited by C. DeWitt *et al.* (Gordon and Breach , New York, 1965).
[20] R. P. Feynman, Phys. Rev. **84**, 108 (1951).
[21] L. Bányai and P. Gartner, Phys. Rev. Lett. **88**, 210404 (2002).
[22] The quantity $g^{(2)}(0)$, measured in Ref. [10] and referred to as the second-order quantum coherence, is related to η by $g^{(2)}(0) = 2 - \eta$.
[23] G. Malpuech *et al.*, Phys. Rev. B **65**, 153310 (2002).
[24] F. Tassone and Y. Yamamoto, Phys. Rev. B **59**, 10 830 (1999).