Time dependence of the ground-state population statistics of condensed microcavity polaritons

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The semiclassical approach consisting of Boltzmann equations for the excited states and the ground state supplemented by a Master equation for the probability distribution of the condensate population is solved for picosecond pulsed microcavity polaritons. With the simple birth- and death-type Master equation for the condensate population that disregards the condensate phase, one gets results, which, compared to quasistationary solutions, are in better agreement with the experiments. First, the time-dependent solutions show the influence of the change to the coherent state already at pump powers close above the condensate threshold. Second, and even more important for the interpretation of corresponding pulsed experiments, the time-dependent solution give results for the second-order correlation function with seemingly larger correlations above threshold, although no polariton-polariton interaction in the ground state has been included.

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I. INTRODUCTION

Many observations on optically excited polaritons in microcavities can consistently be described in terms of a nonequilibrium finite-size Bose-Einstein condensation (BEC).[1,2] In these structures, one of the lowest photon modes is in resonance with excitons confined in one or several quantum wells. The collective excitations of these systems are polaritons. In particular, the lower polariton branch is well separated from the higher branch by a gap that is larger than the damping of the polaritons (strong coupling case). The excited polaritons relax by phonon and particle-particle scattering to the ground state. At sufficiently low temperatures and sufficiently strong pumping, a condensation in the ground state forms, which is directly observable in terms of an onset of laser action of the lowest photon mode of the cavity. An important test of the coherence properties of the condensate is the temporal intensity-intensity correlation function that has been of crucial importance for characterizing the coherence properties of laser light. Measurements of this function have already been performed by Deng *et al.* for pulse-excited microcavities (mc's).[3] Similar experiments have been performed with CdTe mc's.[4] A first attempt to explain these measurements is due to Laussy *et al.*[5] in terms of a coupled Boltzmann equation and Master equation kinetics. Successively, we used the same approach consisting of the coupled Boltzmann-Master equations but with both the polariton-phonon and the polariton-polariton scattering to calculate the second-order correlation function for GaAs mc's under quasistationary excitation.[6] It has been found that the observed large correlations can be understood in quasiequilibrium only if one includes remarkably strong two-quantum scattering processes, as first recognized by Schwendimann and Quattropani.[7] In contrast to Ref. 6, we determine here the correlations for picosecond pulsed excitation.

II. BOLTZMANN-MASTER EQUATIONS

In this section, we recapitulate briefly the structure of the Boltzmann condensation kinetics of the mc polaritons in the lower branch. We mention again that we take the finite cross-section of the mc for the two-dimensional polaritons into account. This finite geometry causes a gap in the energy spectrum between the ground state and the first excited states. For this reason, one obtains at finite temperatures a finite size nonequilibrium Bose-Einstein condensation. For the relaxation kinetics, we take the polariton-polariton (p-p) and the polariton-phonon (p-ph) interactions into account. The structure of the Boltzmann equations for the excited state population $n_k(t)$ is

$$\frac{\partial}{\partial t}n_{\vec{k}} = P_{\vec{k}}(t) - \frac{n_{\vec{k}}}{\tau_{\vec{k}}} + \left.\frac{\partial}{\partial t}n_{\vec{k}}\right|_{p-p} + \left.\frac{\partial}{\partial t}n_{\vec{k}}\right|_{p-\mathrm{ph}}. \tag{1}$$

Here, $P_{\vec{k}}(t)$ is the time-dependent pump rate

$$P_{\mathrm{veck}}(t) = P_0 e^{-\hbar^2(k-k_p)^2/2m_x\Delta E} e^{-2\ln 2 t^2/t_p^2}, \tag{2}$$

where the pump wave number $k_p = 1.7\times 10^5$ cm^{-1} has been chosen to be well above the resonance where coherent scattering into the ground state becomes possible. The energy spread of the pump range is assumed to be $\delta E = 0.1$ meV. The pump duration t_p is assumed here to be 2.5 ps.

The inverse polariton lifetime $1/\tau_{\vec{k}}$ is decomposed into the large mc photon decay rate and the smaller one of the quantum well exciton each weighted by the square of the corresponding Hopfield coefficients. The p-p scattering rates have the basic form

$$\left.\frac{\partial}{\partial t}n_{\vec{k}}\right|_{p-p} = -\sum_{\vec{k}',\vec{k}_1,\vec{k}_2} w^{p-p}_{\vec{k},\vec{k}',\vec{k}_1,\vec{k}_2}\left[n_{\vec{k}}n_{\vec{k}'}(1+n_{\vec{k}_1})(1+n_{\vec{k}_2}) - n_{\vec{k}_1}n_{\vec{k}_2}(1+n_{\vec{k}})(1+n_{\vec{k}'})\right], \tag{3}$$

where $\vec{k}_1 = \vec{k}+\vec{q}$ and $\vec{k}_2 = \vec{k}'-\vec{q}$. The polariton scattering rate for emission and absorption of an acoustic phonon is

$$\left.\frac{\partial}{\partial t}n_{\vec{k}}\right|_{p-\mathrm{ph}} = -\sum_{\vec{q},\sigma=\pm 1} w^{p-\mathrm{ph}}_{\vec{k},\vec{q},\sigma} \times [n_{\vec{k}}(1+n_{k\mp q})N_{q,\sigma} - n_{\vec{k}+\vec{q}}(1+n_{\vec{k}})N_{q,-\sigma}], \tag{4}$$

where the initial and final state phonon numbers are $N_{q,\sigma}=N_q+\frac{1}{2}+\frac{\sigma}{2}$, with the thermal Bose distribution $N_q=[\exp(\hbar\omega_q\beta)-1]^{-1}$. The transition probability is determined by the deformation potential coupling of an electron-hole pair to the lattice. If we solve the Boltzmann equations for $n_k(t)$ and $n_0(t)$ for a short ps pump pulse, one finds that about 20–30 ps after the excitation pulse a local equilibrium distribution is established. The distribution over the excited states can be described by a Bose-Einstein distribution with a rather small degeneracy parameter μ/kT and a temperature close to the bath temperature. The Boltzmann equation (1) for the ground-state population can be written in terms of the scattering rates in and out of the ground-state R_{in} and R_{out}, respectively

$$\frac{\partial\langle n_0\rangle}{\partial t} = \langle R_{\mathrm{in}}(t)(1+n_0(t))\rangle - \langle R_{\mathrm{out}}(t)n_0(t)\rangle. \tag{5}$$

The rate-in is given by the scattering processes from the excited states to the ground state by polariton-polariton $(p-p)$ and polariton-phonon $(p-\mathrm{ph})$ scattering

$$R_{\mathrm{in}} = \sum_{k,k'} w^{p-p}_{0,k;k',k-k'}(1+n_k)n_{k'}n_{k-k'} + \sum_{q,\sigma=\pm 1} w^{p-\mathrm{ph}}_{0,q\sigma}n_q N_{q,-\sigma}. \tag{6}$$

The rate-out is given by

$$R_{\mathrm{out}} = \frac{1}{\tau_0} + \sum_{k,k'} w^{p-p}_{0,k;k',k-k'}n_k(1+n_{k'})(1+n_{k-k'}) + \sum_{q,\sigma=\pm 1} w^{p-\mathrm{ph}}_{0,q\sigma}(1+n_q)N_{q,-\sigma}. \tag{7}$$

Because one has to calculate for the second-order correlation function second moments of the condensate population, we need a stochastic extension of the kinetic equation for the condensate. With the generation $G_n=R^{\mathrm{in}}_n(n+1)$ and the decay rate $D_n=R^{\mathrm{out}}_n n$, we get the Master equation

$$\frac{dW_n}{dt} = -(G_n+D_n)W_n + D_{n+1}W_{n+1} + G_{n-1}W_{n-1}. \tag{8}$$

The rate equation does not provide the full information for the construction of the corresponding Master equation, because the transition rates are only known for $\langle n_0\rangle$ but not for arbitrary $n=n_0$.[11] A simple procedure to include this correction has been formulated by Laussy *et al.*[5] by expanding the distributions of the excited states linearly $n_k(n)=\langle n_k\rangle+\frac{\partial n_k}{\partial n}(n-\langle n\rangle)$ in the deviation of the ground-state population n around its mean value $\langle n\rangle$, and estimating $\frac{\partial\langle n_k\rangle\langle n\rangle}{\partial n}|_{\langle n\rangle} \simeq \frac{\langle n_k\rangle}{\langle n\rangle-N}$, where N is the total number of polaritons. This yields $\langle n_k\rangle(n)\simeq\langle n_k\rangle(1+\frac{n-\langle n\rangle}{\langle n\rangle-N})$. One sees that these correction terms of the scattering rates in the Master equation vanish approximately, if one calculates with the modified Master equation and with $\langle n\rangle=\Sigma_n nW_n$ the rate equation again. We will use these corrections of the scattering rates in linear approximation.

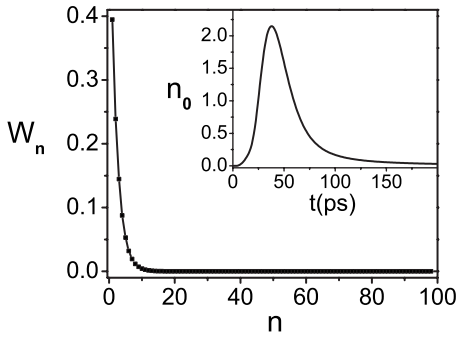

FIG. 1. Calculated distribution functions W_n for a pumping below threshold $P/P_{\mathrm{th}}=0.9$ at the time $t=50$ ps after the pulse. The inset shows the temporal evolution $n_0(t)$

III. SOLUTIONS FOR PULSED EXCITATIONS

The solutions of the Boltzmann-Master equations under quasistationary conditions[6] gave second-order correlation functions which deviated in two important aspects from the experimental observations. The decay from the thermal limit $g^{(2)}=2$ below threshold occurred above threshold only very gradually, but it reached the coherent limit already at twice the pump power. It seems that in the semiclassical description the transition to the coherent phase sets in with increasing pump power only in a retarded way. But in the experiment, the decay of $g^{(2)}$ never reaches the ideal coherent limit of 1. The later fact indicates stronger correlations in the polariton condensate compared to, e.g., the photons in a coherent laser state. Schwendimann and Quattropani[7] used the interaction of two ground-state polariton with two excited polariton states which produced the larger correlations above threshold. However, as this process is nonresonant, it remains questionable[6] whether this process alone can explain the observed large correlations above threshold. Other recent investigations of the second-order correlation used the analogy with atomic lasers[8,9] or used a Langevin fluctuation theory.[9,10]

We will show by self-consistent time-dependent solutions of the mean-field Boltzmann distributions $n_k(t), n_0(t)$ and the probability distribution of the ground-state population $W_n(t)$ that at least for pulsed excitations both discussed deficiencies are no longer as severe as for the stationary solutions. Thus, one needs at least considerably less additional correlations in the condensate to explain the experiments. Laussy *et al.*[5] have also given time-dependent solutions for a gradually switched-on pump, which leads after some time to a stationary state. Rubo *et al.*[12] showed in the treatment of a polariton amplifier an interesting way to go beyond the Boltzmann-Master equation by replacing the semiclassical Master equation by an equation for the quantum statistical operator. In order to get a coherent amplitude, they used a dynamical symmetry-breaking in terms of a small coherent-state initial condition for the statistical operator.

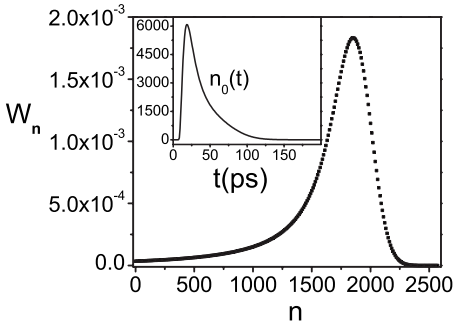

FIG. 2. Calculated distribution functions W_n for a pumping above threshold $P/P_{\mathrm{th}}=2.5$ at the time $t=50$ ps after the pulse. The inset shows the temporal evolution $n_0(t)$

In accordance with the experiment, we use a zero detuning and the Gaussian 2.5-ps pump pulse (2), otherwise all parameters are those of Ref. 6 for GaAs microcavities. We would like to mention that the above described Boltzmann relaxation kinetics (alone, i.e., without the coupled Master equation) with polariton-polariton and polariton-acoustic phonon scattering yielded about 20 ps after a 3-ps pump pulse a thermalization of the excited polariton states. After thermalization, the polariton distribution can be fitted by a local equilibrium Bose-Einstein distribution with slowly varying time-dependent temperature and chemical potential.[13] The calculated thermalization and relaxation scenario is in excellent agreement with corresponding experiments,[14] in particular its strong dependence on the detuning between the photon and exciton modes. Both experiment and numerical studies showed that the polariton gas temperature T relaxes toward the lattice temperature T_l and that the chemical potential μ remains for a relatively long time in the quantum degenerate region where $\mu/kT \ll 1$. The additionally coupled Master equation considered here does not change these results.

We first show in Figs. 1 and 2 the ground-state population distribution $W_n(t=50$ ps) after the pump pulse together with the temporal evolution of the mean ground-state population $n_0(t)$ below but close to threshold and above threshold $P/P_{\mathrm{th}}=2.5$. Note the different scales for $n_0(t)$ below and above threshold. Both results resemble qualitatively those obtained in the quasistationary situation. Because one has to

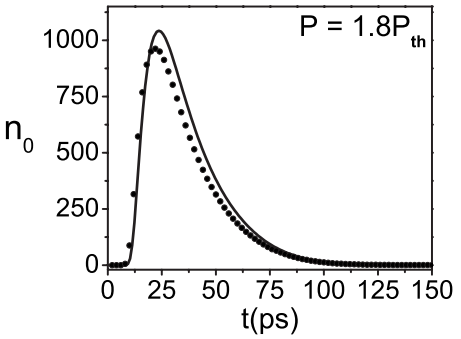

FIG. 3. Mean condensate population as obtained from the Boltzmann equations (full line) and from the solution of the Master equation with $n_0(t)=\Sigma_n n W_n(t)$ (circles)

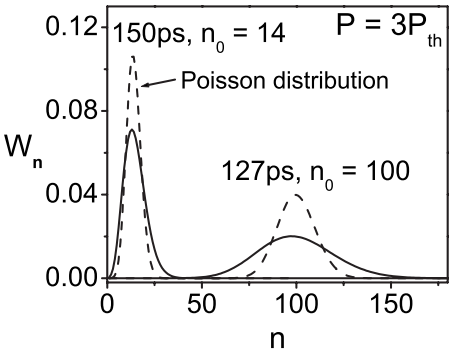

FIG. 4. Calculated probability distribution function $W_n(t=127$ ps) and $W_n(t=150$ ps) for a pump power $P/P_{\mathrm{th}}=3$. For comparison, the Poisson distribution (dashed line) is shown with the same mean condensate population

include a gain saturation in the Master equation, one has to convince oneself, that the mean ground-state population obtained directly from the Boltzmann equation and that obtained with the solution of the Master equation $n_0(t)=\Sigma_n n W_n(t)$ are in reasonable agreement. Figure 3 shows that both results agree reasonably well, if one considers the rough estimate of the gain saturation.[5]

Next, we will show in Fig. 4 that the time-dependent solutions for $W_n(t)$ deviate even three times above threshold considerably from an ideal Poisson distribution with the same mean number of polaritons in the ground state. 127 ps after the pulse, the mean polariton number is 100. It is seen that the actual distribution is considerably broader than a corresponding Poisson distribution, although we know that the system is in a quasithermal equilibrium for times larger than about 40 ps. 150 ps after the pulse, a qualitative similar relation exists, although the decay processes reduced the mean number of condensed polaritons already to 14. The time-dependent second-order population correlation at zero delay $\tau=0$ is

$$g^{(2)}(\tau=0,t)=\frac{\Sigma_n n(n-1)W_n(t)}{\left[\Sigma_n n W_n(t)\right]^2}. \tag{9}$$

In Fig. 5, $g^{(2)}$ is plotted in the time range of 0–200 ps for normalized pump powers between 0.9 and 2.5. In the experiment, only a $\overline{g^{(2)}} \simeq \int_0^T \frac{dt}{T} g^{(2)}(0,t)$ averaged over the whole time interval T of the optical output. In Fig. 6, the averaged

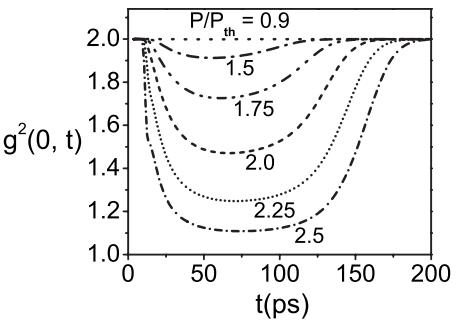

FIG. 5. Calculated second-order coherence function $g^{(2)}(0,t)$ versus t for various normalized pump powers

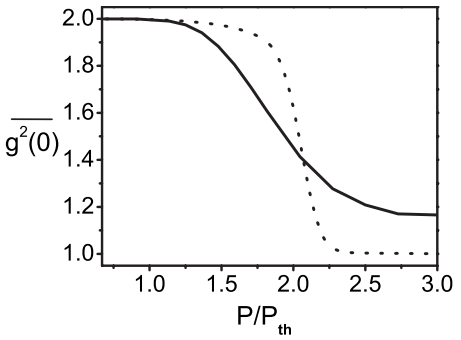

FIG. 6. Calculated time-averaged second-order coherence function $\overline{g^{(2)}}(0)$ versus normalized pump power. The dotted line gives the stationary results of Ref. 6

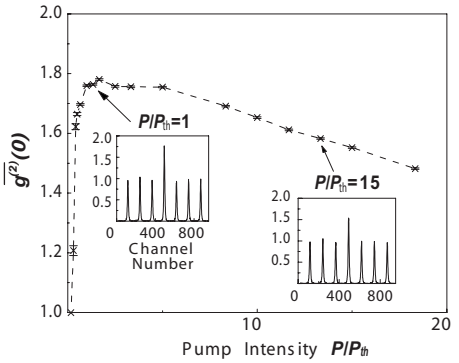

FIG. 7. Measured time-averaged second-order coherence function $g^{(2)}(0)$ versus normalized pump power P/P_{th} according to Deng *et al.* (Ref. 3) for GaAs mc's

second-order correlation function is compared to the quasistationary one for normalized pump powers up to 3. One sees that the full time-dependent solution for the distribution function $W_n(t)$ yields a correlation function whose decay starts right at threshold. The fast transients obviously help to trigger the transition from the thermal state to the coherent state which however is in the available finite life time of the mc polaritons not reached in its ideal Poisson limit (see Fig. 4). At the same time, Fig. 6 shows that $\overline{g^{(2)}}$ decreases slower with increasing pump power than the quasistationary $g^{(2)}$.[6] Particularly important is that $\overline{g^{(2)}(P/P_{\mathrm{th}})}$ levels off at a correlation larger than the ideal coherent value of one. Thus the time-dependence alone causes a fictitious correlation in the condensate, although we did in the present calculations not take into account any polariton-polariton interaction in the ground state. The described features of the averaged correlation function brings it closer to the experimental results[3] reproduced in Fig. 7. It should be remarked that the measurement of the second-order correlation function suffers from limited time resolution in the threshold region, so the thermal limit of two is not reached.

In conclusion, it is shown that the need to consider additional interaction effects between polaritons in the condensate is by the time-dependent solutions of the Boltzmann-Master equations—at least for experiments that are not truly stationary—considerably reduced. For the same excitation conditions where we get improved $g^{(2)}$ values, the considered kinetics yields polariton distributions[13] that have been found to be in very good agreement with the experiment.[14]

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