Coherence of condensed microcavity polaritons calculated within Boltzmann-Master equations

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The first-order spatial and the second-order temporal coherences of condensed microcavity polaritons are calculated in terms of Boltzmann equations for the excited states and the ground state supplemented by a Master equation for the probability to find a given number of particles in the condensate. The resulting first-order spatial coherence agrees both in its pump power dependence and in its variation with distance with the results of a recent double-slit experiment. Inserting the calculated rates between the excited states and the condensate with various saturation and depletion models, we solve for stationary situations the Master equation for the ground-state population. The resulting second-order correlations for the various models are compared with recent measurements.

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I. INTRODUCTION

In recent years a nonequilibrium finite-size Bose-Einstein condensation (BEC) has been observed in optically excited semiconductor microstructures.[1,2] In these structures the photons are confined by a microcavity (mc) with Bragg mirrors on both sides, the photons are in resonance with excitons confined in one or several quantum wells. The collective excitations of these systems are polaritons. In particular the lowest branch is well separated from the next-higher branch by a gap which 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 occurs which is directly observable in terms of an onset of laser action of the lowest photon mode of the cavity. The ease of observation of the condensation in terms of a connected laser emission generated also some doubts whether on can really speak about a nonequilibrium BEC of polaritons or whether on has just realized another exciton laser. Other observed features added more and more evidence to the interpretation in terms of a BEC. Examples are (i) the observation of a thermal, degenerate Bose-Einstein distribution of the excited polaritons[3] in agreement with corresponding treatments of the polariton kinetics,[4] (ii) the observation of a stable spin polarization of the condensed state[1] as predicted by a quasispin kinetics,[5,6] and very importantly the direct observation of the Bogoliubov spectrum in angle-resolved luminescence in new GaAs mc's with lifetimes as long as 80 ps.[7]

One of the most direct evidence of a macroscopic condensate is the observation of coherence. First-order spatial coherence has been detected recently in a double-slit experiment.[8] An early attempt to calculate the first-order spatial correlation function is due to Sarchi and Savona.[9] With their condensation kinetics they obtained relatively large condensate densities which resulted in a too strong off-diagonal long-range order.[10] They incorporated many-body effects using the Popov approximation in order to get a depletion of the condensate. We will later see that at least for the GaAs mc's with a 2 ps lifetime our kinetics yields condensate fractions which give the observed off-diagonal spatial coherence,[8] which leaves not much space to larger depletion effects.

The first-order temporal coherence function yields the linewidth of the polariton laser light. Interestingly, the linewidth above threshold does not decrease according to the Shallow-Townes formula but increases again[11,12] with increasing density of the polaritons in the ground state. This observed effect can be calculated by Langevin equations for the Bogoliubov model of weakly interacting bosons in terms of an interaction of the polaritons in the ground state which gives rise to a Kerr-effect-like behavior which in turns causes the linewidth increase.

Another important test of the coherence properties of the condensate is the temporal intensity-intensity correlation function which 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.*[14] for GaAs mc's with 2 ps lifetime and recently for GaAs mc's with longer lifetimes (Ref. 15) and by Kasprazk *et al.*[16] in CdTe mc's. A first attempt to explain these measurements is due to Laussy *et al.*[18] in terms of a coupled Boltzmann equation and Master equation kinetics.

Here we will investigate in Sec. II to which extent the observation of the spatial coherence in the 2 ps GaAs mc's can be explained in terms of a semiclassical Boltzmann kinetics taking into account both polariton-phonon and polariton-polariton scatterings. In the following section we will show that one can calculate directly the spatial variation in the first-order coherence function and its dependence on the pump power by using the distributions obtained from the Boltzmann kinetics. We perform a detailed comparison with the measured variations with pump power and distance in 2 ps GaAs-type mc's.[8]

Using a stochastic extension of the Boltzmann kinetics, we treat in Sec. III the coupled Boltzmann equations and the Master equation for the probability to find n polaritons in the

ground state. We include gain saturation and interactions which deplete the condensate. For stationary situations we solve the Master equation iteratively. The second-order correlation function $g^{(2)}$ which can be calculated with the solutions of the Master equation show for various saturation or depletion models bunching of condensed polaritons above threshold. The features of the calculated $g^{(2)}$ for the various saturation or depletion models are compared with the measured ones in GaAs mc's by Roumpos *et al.*[15] In order to explain the slow decay of correlations with increasing pumping seen in GaAs mc's (Ref. 15) and also in CdTe mc's,[16] one has to include remarkably strong two-quantum scattering processes as also observed recently by Schwendimann and Quattropani.[17]

II. FIRST-ORDER SPATIAL COHERENCE

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 (2D) 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 (BEC). For the relaxation kinetics we take the polariton-polariton (p-p) and the polariton-phonon (p-ph) interactions into account. For details of the model and material parameters for the GaAs-type mc's we refer to our earlier publications.[4,22] The structure of the Boltzmann equations for the distribution in the excited states $n_k(t)$ and in the ground state $n_0(t)$ is

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

$$\frac{\partial}{\partial t}n_0 = -\frac{n_0}{\tau_0} + \left.\frac{\partial}{\partial t}n_0\right|_{\text{p-p}} + \left.\frac{\partial}{\partial t}n_0\right|_{\text{p-ph}}. \tag{2}$$

Here, $P_{\vec{k}}(t)$ is the time-dependent pump rate and $\tau_{\vec{k}}$ is the polariton lifetime. The p-p scattering rates have the basic form

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

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

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

where the initial and final-state phonon numbers are $N_{q,\sigma}=N_q+\frac{1}{2}+\frac{\sigma}{2}$, with the phonon Bose distribution $N_q=\frac{1}{e^{\beta_l\hbar q v_s}-1}$, where β_l is the inverse thermal lattice energy and v_s is the sound velocity. The transition probability is determined by the deformation-potential coupling of an electron-hole pair to the lattice. If we solve for a given pump pulse the Boltzmann equations for $n_k(t)$ and $n_0(t)$ about 20–30 ps after the excitation pulse a local equilibrium distribution is established.[4] 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. In the following we use all parameter of Ref. 4, in particular we use a positive detuning of 4 meV and a cross section of 100 μm^2. Both parameters are known to result in a relatively fast condensation kinetics.

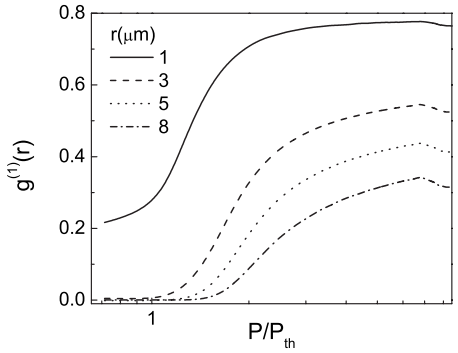

FIG. 1. Calculated first-order coherence function $g^{(1)}(r)$ versus normalized pump power P/P_{th} for various distances r.

With these solutions we will now calculate the first-order spatial coherence function defined as

$$g^{(1)}(\vec{r}_1,\vec{r}_2) = \frac{\langle\psi^\dagger(\vec{r}_1)\psi(\vec{r}_2\rangle}{\langle\psi^\dagger(\vec{r}_1)\rangle\langle\psi(\vec{r}_2)\rangle}. \tag{5}$$

Inserting a plane-wave expansion

$$\psi(\vec{r}) = \frac{1}{\sqrt{S}}\left(b_0 + \sum_{\vec{k}} b_k e^{i\vec{k}\cdot\vec{r}}\right), \tag{6}$$

we find in the free-particle approximation

$$g^{(1)}(\vec{r}_1,\vec{r}_2) = \frac{n_0 + \sum_{\vec{k}} n_k e^{i\vec{k}\cdot(\vec{r}_1-\vec{r}_2)}}{n_0}. \tag{7}$$

With the calculated populations we can now evaluate the pump power dependence of $g^{(1)}(r=|\vec{r}_1-\vec{r}_2|)$ for various values of the distance r. The pump power P is normalized to the threshold value P_{th}. One sees clearly from Fig. 1 how the range of the spatial coherence increases above threshold. These results are in excellent agreement of those measured with a double-slit experiment by Deng *et al.*[8] shown in Fig. 2. As a next test we plot the distance dependence of $g^{(1)}(r)$ in Fig. 3 for various values of the pump power. We see a more or less exponential decay of the coherence function. The coherence length increases with pump power, i.e., with the degeneracy of the condensed polaritons. In Fig. 4 we show again the corresponding measurements of Ref. 8 which are again in nearly quantitative agreement with the calculated values of the coherence function.

Critically one should remark in this context that there is some ambiguity in the modeling of the condensation kinetics in 2D systems. In order to get a finite-size condensation in

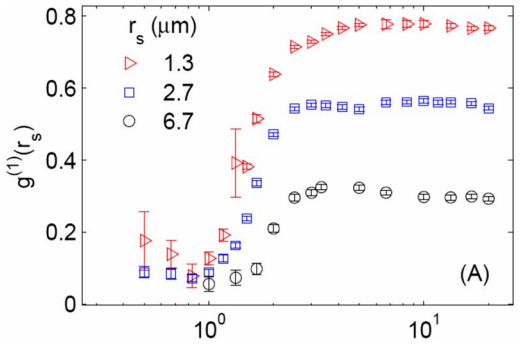

FIG. 2. (Color online) Measured first-order coherence function $g^{(1)}(r)$ versus normalized pump power P/P_{th} for various distances r according to Deng *et al.* (Ref. 8).

2D, one has to assume a finite cross section of the mc's which, however, is not known very well. It is often not given by the geometry of the device, but by the spot size of the exciting laser. A small cross section results in a large energy gap between the ground state and the first-excited states, and thus in a strong condensation with a large condensate density. In this respect we used the 100 μm^2 cross section simply because our earlier experience showed us that with this choice we get a rather good description of the condensation kinetics in terms of the Boltzmann equation in the 2 ps GaAs mc's (Ref. 4) as mentioned above. It is too early to decide whether a somewhat smaller cross section in combination with a quantum kinetics—which would result some depletion—is more appropriate. Because the interactions are stronger in II–VI compound mc's, this question is even more urgent in these systems.[9]

III. INTENSITY-INTENSITY CORRELATION FUNCTION

In order to test the coherence properties of a polariton condensate, one has to perform a Hanbury Brown-Twiss experiment in which the correlations of the intensity $I(t+\tau)$ at time $t+\tau$ and the intensity $I(t)$ at time t are measured by the second-order correlation function which is in its classical form

$$g_{\text{cl}}^{(2)}(\tau) = \frac{\langle I(t+\tau)I(t)\rangle}{\langle I(t+\tau)\rangle\langle I(t)\rangle}. \tag{8}$$

The intensity of the polariton laser light is directly proportional to the population of the ground state in which the

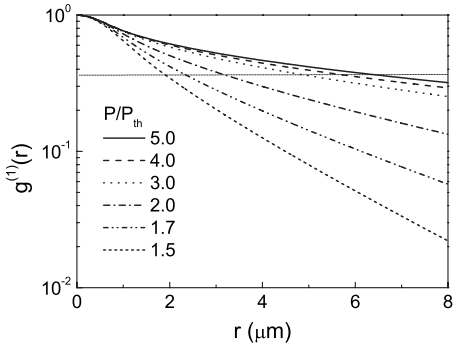

FIG. 3. Calculated first-order coherence function $g^{(1)}(r)$ versus distances r for various normalized pump powers P/P_{th}.

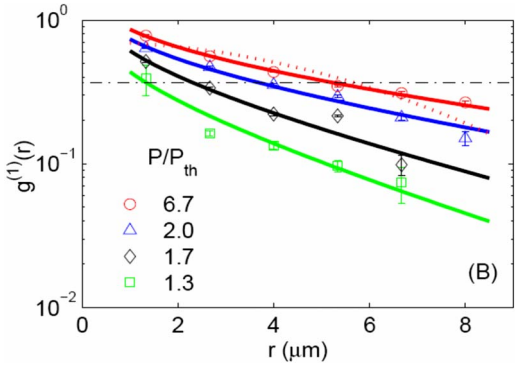

FIG. 4. (Color online) Measured first-order coherence function $g^{(1)}(r)$ versus distances r for various normalized pump powers P/P_{th} according to Deng *et al.* (Ref. 8).

polaritons condense above threshold. The quantum-statistical correlation function $g^{(2)}$ is given by the correlations of the condensate operators[23]

$$g^{(2)}(\tau) = \frac{\langle b_0^\dagger(t)b_0^\dagger(t+\tau)b_0(t+\tau)b_0(t)\rangle}{\langle b_0^\dagger(t+\tau)b_0(t+\tau)\rangle\langle b_0^\dagger(t)b_0(t)\rangle}. \tag{9}$$

The mean condensate density $\langle n_0(t)\rangle = \langle b_0^\dagger(t)b_0(t)\rangle$ is determined by a Boltzmann Eq. (2) which governs the scattering kinetics in and out of the ground state

$$\frac{\partial\langle n_0\rangle}{\partial t} = \langle R_{\text{in}}(t)[1+n_0(t)]\rangle - \langle R_{\text{out}}(t)n_0(t)\rangle. \tag{10}$$

The rate in is given by the scattering processes from the excited states to the ground state by polariton-polariton and polariton-phonon scattering

$$R_{\text{in}} = \sum_{k,k'} w_{0,k;k',k-k'}^{\text{p-p}}(1+n_k)n_{k'}n_{k-k'} + \sum_{q,\sigma=\pm1} w_{0,q\sigma}^{\text{p-ph}}n_q N_{q,-\sigma}. \tag{11}$$

The rate out is given by

$$R_{\text{out}} = \frac{1}{\tau_0} + \sum_{k,k'} w_{0,k;k',k-k'}^{\text{p-p}}n_k(1+n_{k'})(1+n_{k-k'}) + \sum_{q,\sigma=\pm1} w_{0,q\sigma}^{\text{p-ph}}(1+n_q)N_{q,-\sigma}. \tag{12}$$

As a comment we mention that formally one has exchange rates due to the scattering process $\vec{k}$, $0\to 0$, $\vec{k}$. These exchange rates do not give raise to changes in the populations and thus should not be included in the kinetics. The mean rate equation can thus be written as

$$\frac{\partial\langle n_0\rangle}{\partial t} = \langle R_{\text{in}}(t)\rangle\langle[1+n_0(t)]\rangle - \langle R_{\text{out}}(t)\rangle\langle n_0(t)\rangle. \tag{13}$$

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. There are at least four formulations of such an extension possible

(A) One can supplement the Boltzmann equation for n_0 by Langevin fluctuations with shot-noise character.[19–21] The second density moments are then determined by the second moments of these shot-noise fluctuations. As it is known from laser theory, Langevin equations can be solved approximately by linearization below and above threshold, but there is no simple way to find solutions in the whole density regime.

(B) This is different for the associate Fokker-Planck equation in which the probability for a certain density value is calculated. In stationary equilibrium, one gets an analytic solution in terms of an exponential of a generalized Ginzburg-Landau potential (see e.g., Risken[24]). The change in the potential from below threshold with a minimum at $n_0=0$ to a potential with a sharp minimum at $n_0 \neq 0$ above threshold describes the probabilities for all densities. In both approaches, the Langevin and the Fokker-Planck method, the density is considered to be a continuous variable which is valid if $n_0 \gg 1$. Because in a BEC the condensate density varies from small values of $n_0 \ll 1$ below threshold to large values above $n_0 \gg 1$ this approximation is not well justified below and at threshold where $n_0=\mathrm{O}(1)$.

(C) The third possible stochastic extension of the ground-state kinetics is the Master equation for the probability to find n_0 particles in this state. The Master equation takes the discrete nature of the number of condensed particles exactly into account.[25] We thus adopt this approach which has been considered before by Laussy *et al.*[18] These authors calculated $g^{(2)}(0)$ for CdTe mc's (using only phonon-polariton and electron-polariton scattering) from numerical solutions of the time-dependent Master equation. In contrast, we will use a well-converging detailed-balance and continued-fraction methods for the solution of the stationary Master equation and calculate the results for GaAs mc's (using both ph-p and p-p scattering) in order to be able to compare the results with Yamamoto's experiment.[14]

A simplification to all three approaches is to limit the treatment of fluctuations only for the condensate, but treat the excited states simply by a mean equation, i.e., the Boltzmann equation. We will discuss below the limitations of this approach.

(D) Still another possibility is to treat the kinetics of the second-order function $\langle b_0^\dagger(t) b_0^\dagger(t) b_0(t) b_0(t) \rangle$ in addition to the kinetics of the densities. This approach has recently be applied by Schwendimann and Quattropani.[17]

IV. MASTER EQUATION

We will now formulate the Master equation for the probability $W_{n_0}(t)$ to find at time t n_0 particles in the condensate. With the generation and decay rate G_{n_0} and D_{n_0}, respectively,

$$G_{n_0} = R_{n_0}^{\mathrm{in}}(n_0+1) \quad \text{and} \quad D_{n_0} = R_{n_0}^{\mathrm{out}} n_0, \tag{14}$$

we get the Master equation

$$\frac{dW_{n_0}}{dt} = -(G_{n_0}+D_{n_0})W_{n_0} + D_{n_0+1}W_{n_0+1} + G_{n_0-1}W_{n_0-1} = \sum_{m_0=n_0-1}^{m_0=n_0+1} M_{n_0,m_0} W_{m_0}. \tag{15}$$

$M_{n,m}$ is a tridiagonal matrix.[24] Its elements are

$$M_{n_0,n_0} = -(G_{n_0}+D_{n_0}), \quad M_{n_0,n_0+1} = D_{n_0+1}, \quad M_{n_0,n_0-1} = G_{n_0-1}. \tag{16}$$

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 $n=n_0$.[25] Systematically, one would have to determine the populations of the excited states as a function of the pump rate and the number of polaritons in the ground state, as it can be done for a homogeneously broaden three-level laser analytically. But, here we know the mean rates from the solution of the Boltzmann equation only numerically. A simple procedure to include the gain saturation has been formulated by Laussy *et al.*[18] by expanding the distributions of the excited states linearly $n_k(n_0)=\langle n_k \rangle + (\partial n_k/\partial n_0)(n_0-\langle n_0 \rangle)$ in the deviation of the ground-state population n_0 around its mean value $\langle n_0 \rangle$. In order to avoid confusions $\langle n_k \rangle$, $\langle n_0 \rangle$ are the solutions of the Boltzmann equations, called only n_k, n_0 above. However, in the stochastic extension of this kinetics, we have to make the distinction between the mean values and the actual values. Estimating the derivative, Laussy *et al.* get

$$\left.\frac{\partial \langle n_k \rangle(n_0)}{\partial n_0}\right|_{\langle n_0 \rangle} = \frac{\langle n_k \rangle}{\langle n_0 \rangle - N}, \tag{17}$$

where N is the total number of polaritons. This yields

$$\langle n_k \rangle(n_0) = \langle n_k \rangle \left(1 + \frac{n_0 - \langle n_0 \rangle}{\langle n_0 \rangle - N}\right). \tag{18}$$

One has to insert these excited-state populations with gain saturation into the gain and decay rates known from the Boltzmann equations. 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 $\langle n_0 \rangle = \Sigma_{n_0} n_0 W_{n_0}$ the rate equation again. We will use these corrections of the scattering rates in linear approximation. Because for all excited states $1 \gg \langle n_k \rangle$, the correction can be neglected for the final-state factors $1+\langle n_k \rangle$. Thus the polariton-phonon scattering rates $R_{\mathrm{in}}^{\mathrm{p\text{-}ph}}$ will get a correction term with the factor $F(n_0)=\frac{n_0-\langle n_0 \rangle}{\langle n_0 \rangle - N}$, while the correction term of polariton-polariton scattering rates in $R_{\mathrm{in}}^{\mathrm{p\text{-}p}}$ get the factor $2F(n_0)$. In the rate out, only the polariton-polariton scattering rates will be modified. The correction term $R_{\mathrm{out}}^{\mathrm{p\text{-}p}}$ gets the factor $F(n_0)$, while the other rates remain unchanged.

A. Stationary solution

We consider stationary pumping. Once a stationary state is reached, the rates in and out of the ground state do no longer change with time but will naturally depend on the pump strength. Thus we can treat the stationary Master equation.

B. Stationary solution obtained from detailed balance

The stationary solution is maintained if the rates between two successive states balance. From Fig. 5 we see that the following detailed-balance relation holds:

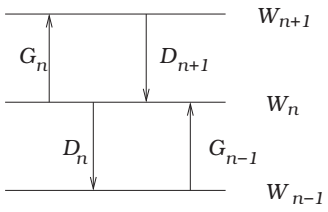

FIG. 5. Level diagram for Master equation.

$$D_{n_0} W_{n_0} = G_{n_0-1} W_{n_0-1}, \tag{19}$$

which yields the solution

$$W_{n_0} = \frac{G_{n_0-1}}{D_{n_0}} W_{n_0-1} = \prod_{k=1}^{k=n_0} \frac{G_{k-1}}{D_k} W_0. \tag{20}$$

W_0 is obtained from the normalization $\Sigma_{n_0} W_{n_0} = 1$.

C. Iterative solution in terms of continued fractions

In general, the stationary solutions are obtained from the equation

$$\sum_{m_0=n_0-1}^{m_0=n_0+1} M_{n_0,m_0} W_{m_0} = 0. \tag{21}$$

Introducing the ratio

$$S_{n_0} = \frac{W_{n_0+1}}{W_{n_0}}, \tag{22}$$

we find

$$M_{n_0,n_0} + M_{n_0,n_0+1} S_{n_0} + \frac{M_{n_0,n_0-1}}{S_{n_0-1}} = 0, \tag{23}$$

which yields the recurrence relation by changing $n_0 \to n_0 + 1$

$$S_{n_0} = -\frac{M_{n_0+1,n_0}}{M_{n_0+1,n_0+1} + M_{n_0+1,n_0+2} S_{n_0+1}}. \tag{24}$$

Equation (24) is well suited for numerical iteration. By analytical iteration one gets the continued fractions for the ratio S_n. Finally, W_n is given by

$$W_{n_0} = S_{n_0-1} S_{n_0-2} \ldots S_0 W_0. \tag{25}$$

The value of W_0 follows again from the normalization condition. One can start the iteration at a value $N \gg \langle n_0 \rangle$ for which one can put $W_{N+1} = W_{N+2} = \ldots = 0$, so that $S_N = 0$. Thus one can calculate $S_{N-1} = -M_{N,N-1}/M_{N,N}$ from Eq. (24), and so on until we get S_0.

V. SECOND-ORDER CORRELATION FUNCTION

The quantum-statistical second-order correlation function for zero delay time is given in terms of field operators b_0 of the condensate according to Eq. (9) by

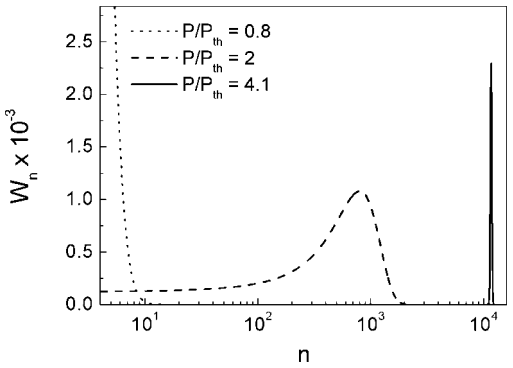

FIG. 6. Calculated distribution functions W_{n_0} for various normalized pump powers P/P_{th}.

$$g^{(2)}(0) = \frac{\langle b_0^\dagger b_0^\dagger b_0 b_0 \rangle}{\langle b_0^\dagger b_0 \rangle \langle b_0^\dagger b_0 \rangle}.$$

Note that the field operators are in the normal form with all creation operators to the right. In a n-diagonal number representation of the density matrix $\rho_{n_0,n_0} = W_{n_0}$ in which n_0 are natural numbers ranging from 0 to the total number of polaritons N, one gets

$$g^{(2)}(0) = \frac{\sum_1^N n_0(n_0-1) W_{n_0}}{\left(\sum_1^N n_0 W_{n_0}\right)^2}, \tag{26}$$

There are well-known limiting cases for $g^{(2)}(0)$: for a thermal distribution $W_{n_0} = \frac{1}{Z}\exp[-\beta(E_0-\mu)n_0]$ with the normalization constant Z, Eq. (26) yields $g^{(2)}(0) = 2$. For a Poisson distribution $W_{n_0} = \frac{\langle n_0 \rangle^{n_0}}{n_0!}\exp(\langle n_0 \rangle)$ which holds in the coherent limit, one gets $g^{(2)}(0) = 1$.

VI. SOLUTIONS OF THE MASTER EQUATION

The stationary probability distributions obtained by iteration are shown in Figs. 6 for three pump powers. Below threshold, the distribution peaks at $n = 0$ and decays monotonically. Slightly above threshold, the distribution peaks at a value $\langle n_0 \rangle \gg 1$ but is still rather broad. Well above threshold

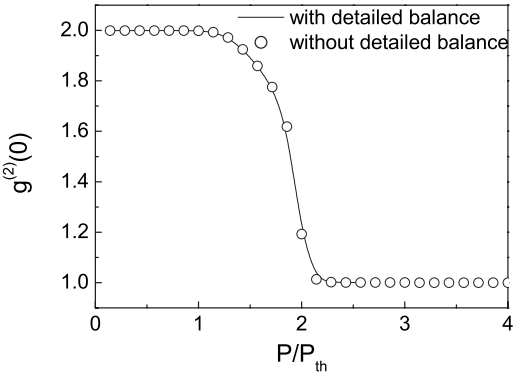

FIG. 7. Calculated second-order coherence function $g^{(2)}(0)$ versus normalized pump power P/P_{th}. The circles give the detailed-balance result.

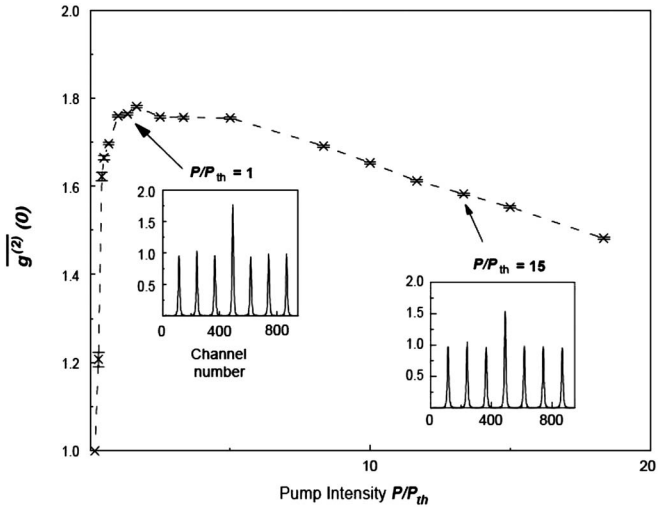

FIG. 8. Measured second-order coherence function $g^{(2)}(0)$ versus normalized pump power P/P_{th} according to Deng *et al.* (Ref. 14) for 2 ps GaAs mc's.

for $P/P_{\mathrm{th}}=4.1$, the distribution peaks sharply around a larger density value of about $\langle n_0\rangle \simeq 10^4$. We checked that the solutions obtained iteratively agree indeed with those obtained from the detailed-balance condition.

Once all W_{n_0} are calculated including the normalization W_0, one gets $g^{(2)}(0)$ from Eq. (26).

We see how the thermal limit with $g^{(2)}(0)=2$ and the coherent limit with $g^{(2)}(0)=1$ are smoothly connected. The transition takes place in the region of pump powers $P \simeq 2P_{\mathrm{th}}$. As will be discussed below this fast decay of the correlations above threshold is not observed in the experiments.

In Fig. 8 we show for comparison the measured $g^{(2)}(0)$ of Ref. 14 measured for 2 p GaAs mc's. Below threshold the average over the rapid fluctuations in the experiment falsifies $g^{(2)}$ so that it decreases below threshold. This can be understood qualitatively, taking into account that $g^{(2)}(\tau)$ varies approximately as $g^{(2)}(\tau)=2e^{(R_{\mathrm{in}}-R_{\mathrm{out}})\tau}$ as one can show, e.g., in a Langevin approach. An average over the delay times yields $\bar{g}_2(0) \propto g^{(2)}(0)/(R_{\mathrm{out}}-R_{\mathrm{in}})$. As the pump power decreases away from threshold $R_{\mathrm{out}}-R_{\mathrm{in}}$ increases. Thus the averaged $\bar{g}_2(0)$ decreases rapidly below threshold, as seen in the experiment with the strongly fluctuating 2 ps GaAs mc's. Above threshold where the influence of time averaging is not so strong, the measured correlations decrease only slowly. In a more recent experiment with GaAs mc's with a longer cavity lifetime, a minimum of $g^{(2)}(P/P_{\mathrm{th}})$ seems to exist at threshold, and the range of correlations above the coherent limes extends over a much wider pump range.[15] In these experiments the strong influence of spectral and angle resolution on the statistical properties of the ground-state emission has been shown. In similar measurements in CdTe mc's[16] even an increase in $g_2(0)$ with pump power above threshold has been found. It seems that other many-body effects have to be included in order to account for the difference between these experimental findings and the theory.

VII. SCATTERING OF TWO CONDENSED POLARITONS

A. Nonresonant polariton-polariton scattering

One way to improve the $g^{(2)}$ calculations is to include two-quanta processes. Following an idea of Schwendimann and Quattropani,[17] one can consider a quadratic correction term in the form of a scattering of two condensate polaritons to the excited states with the momenta q and $-q$. This process is nonresonant, thus only with finite line width this scattering to low-lying excited states can contribute. The transition rates of this process are

$$\sum_q w^{\mathrm{p\text{-}p}}_{0,0;q,-q}[n_0(n_0-1)(1+n_q)(1+n_{-q}) - (1+n_0)(2+n_0)n_q n_{-q}] \tag{27}$$

with $n_0-1\geq 0$. The energy-conserving delta function has to be replaced by a Lorentzian or Gaussian resonance

$$2\pi\delta(2e_0-2e_q) \Rightarrow \frac{2\gamma}{[2(e_0-e_q)]^2+\gamma^2} \quad \text{or} \quad \Rightarrow \frac{2}{\gamma}e^{-4(e_0-e_q)^2/\gamma^2}. \tag{28}$$

This yields two-particle transition rates of the form

$$G^2_{n_0} = R^{\mathrm{in},(2)}(n_0+1)(n_0+2)$$

$$\text{with } R^{\mathrm{in},(2)} = \sum_q w^{\mathrm{p\text{-}p}}_{0,0;q,-q} n_q n_{-q} \tag{29}$$

and

$$D^2_{n_0} = R^{\mathrm{out},(2)} n_0(n_0-1)$$

$$\text{with } R^{\mathrm{out},(2)} = \sum_q w^{\mathrm{p\text{-}p}}_{0,0;q,-q}(1+n_q)(1+n_{-q}). \tag{30}$$

The last finite decay rate is $D_2^2=R^{\mathrm{out},(2)}2$, while $D_1^2=D_0^2=0$. The simple reason is, one needs at least two polaritons in the ground state for the considered quadratic scattering process out of the ground state.

The two quadratic terms extend the three-diagonal to a five-diagonal Master equation

$$\frac{dW_{n_0}}{dt} = -(G_{n_0}+D_{n_0}+G^{(2)}_{n_0}+D^{(2)}_{n_0})W_{n_0} + D_{n_0+1}W_{n_0+1} + D^{(2)}_{n_0+2}W_{n_0+2} + G_{n_0-1}W_{n_0-1} + G^{(2)}_{n_0-2}W_{n_0-2} = \sum_{m_0=n_0-2}^{m_0=n_0+2} M_{n_0,m_0}W_{m_0}. \tag{31}$$

B. Kinetic saturation model for a dense condensate

It is known that 2D excitons eventually ionize once their density is so high that the exciton wave functions start to overlap.[13] The critical saturation density due to phase-space filling has been evaluated from the reduction in the exciton

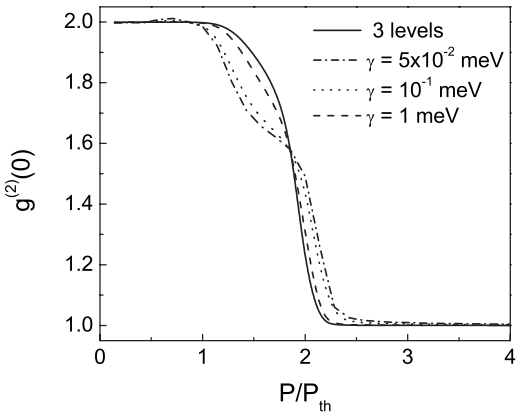

FIG. 9. Calculated second-order coherence function $g^{(2)}(0)$ versus normalized pump power P/P_{th} for the nonresonant two-polariton scattering.

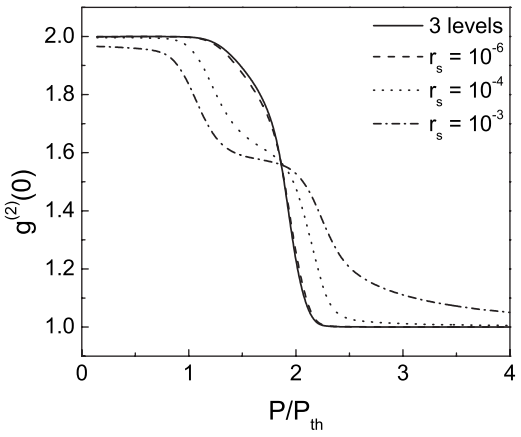

FIG. 10. Calculated second-order coherence function $g^{(2)}(0)$ versus normalized pump power P/P_{th} for the kinetic saturation model.

oscillator strength to be $\frac{1}{n_s}=\frac{32}{7}\pi a_{2\mathrm{D}}^2$. This will also result in a reduction in the exciton density n to $n_0/[1+(n_0/n_s)^2]$. In order to incorporate the saturation effect kinetically in the Master equation, we introduce phenomenologically a two-polariton ionization rate. Due to the overlap of the wave functions in a collision of two exciton polaritons, the two polaritons are assumed to by ionized and thus lost for the condensate

$$D_{n_0}^2=\frac{1}{\tau_0}n_0(n_0-1)\frac{r_s}{n_s}. \tag{32}$$

We have included a small numerical factor r_s in order to avoid an unphysically large ionization rate far below saturation. We used the lifetime τ_0 to set the scale for the ionization rate. Any direct back scattering from the ionization continuum to the condensate can be neglected.

Solution of a five-diagonal Master equation by iteration. For the simultaneous presence of one and two-quanta transitions detailed balance does no longer hold. The stationary Master equation with two-quanta transitions

$$\frac{dW_{n_0}}{dt}=\sum_{m=-2}^{m+2}M_{n_0,n_0+m}W_{n_0+m}=0 \tag{33}$$

will therefore be solved by an iteration procedure starting with a highest nonzero value for W_N, i.e., all $W_{n_0}=0$ for all $n_0\geq N+1$. Thus the Master equation for W_N reads

$$W_N=-\frac{M_{N,N-1}}{M_{N,N}}W_{N-1}-\frac{M_{N,N-2}}{M_{N,N}}W_{N-2}. \tag{34}$$

As shown in detail in the Appendix, we generate all W_{n_0} downward until we express W_1 and thus all higher W_{n_0} by W_0. W_0 is finally determined by the normalization. We show the resulting second-order correlation functions both for the ionization model and for the nonresonant two-polariton scattering out of the condensate. In comparison with the results of Schwendimann and Quattropani[17] which are at least qualitatively in better agreement with the experiments, we find for the nonresonant two-polariton scattering with reasonable collision broadenings a less extended region of increased correlations. Another difference is that they get a relative minimum in the threshold region—at least in their model with an exciton reservoir—which is not present in Fig. 9. In a recent mean-field treatment[26] of the polariton kinetics—similar as ours—the minimum of $g_2(0)$ at threshold as a function of the pump power is nearly absent, resembling in the respect more our results. On the other hand, our treatment is not fully self-consistent because we assumed that the corrections due to the nonresonant quadratic interactions are small for the solution of the mean Boltzmann equation. More importantly, by treating only the ground-state population stochastically, we miss correlations beyond the mean-field treatment between the ground state and excited states at k and $-k$, which may be better accounted for in the treatment of Ref. 17. Another possible effect which can only be obtained by a full stochastic treatment of both the ground and the excited states are relaxation oscillations which have been observed for lasers between the photon number and the populations of the gain medium and which might exist also for condensed mc polaritons.

As an attempt to increase the range of correlations within our approach, we plot in Fig. 10 the results of the kinetic saturation model. We see that with increasing interaction strength r_s the range of correlations where $g^{(2)}\geq 1$ extends to larger pumping rates as it is observed in the experiment. In this general aspect the simple model brings the results closer to those of the experiment than those of Fig. 7.

In conclusion, we have shown that the measured first-order spatial correlations can be explained excellently by a Boltzmann condensation kinetics. The observed large second-order correlations above the coherent limit[14–16] have been studied by combining the Boltzmann equation with the Master equation for the number of condensed particles. Only if a considerable—not fully understood—interaction between the condensed particles is assumed, the experimental results can be understood within our approach. The limitations of treating only the ground-state population stochastically are discussed.

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APPENDIX: ITERATIVE SOLUTION OF FIVE-DIAGONAL MASTER EQUATION

The stationary Master equation with two-quanta transitions

$$\frac{dW_{n_0}}{dt} = \sum_{m=-2}^{m+2} M_{n_0,n_0+m} W_{n_0+m} = 0 \tag{A1}$$

will be solved by an iteration procedure starting with a highest nonzero value for W_N, i.e., all $W_{n_0}=0$ for all $n_0 \geq N+1$. Thus the Master equation for W_N reads

$$W_N = a_N W_{N-1} + b_N W_{N-2}$$

with the coefficients

$$a_N = -\frac{M_{N,N-1}}{M_{N,N}}, \quad b_N = -\frac{M_{N,N-2}}{M_{N,N}}.$$

Substituting this result into $\frac{dW_{N-1}}{dt}=0$, one gets

$$W_{N-1} = a_{N-1} W_{N-2} + b_{N-1} W_{N-3}$$

with the coefficients

$$a_{N-1} = -\frac{M_{N-1,N} b_N + M_{N-1,N-2}}{M_{N-1,N} a_N + M_{N-1,N-1}},$$

$$b_{N-1} = -\frac{M_{N-1,N-3}}{M_{N-1,N} a_N + M_{N-1,N-1}}.$$

Next we substitute this result into $\frac{dW_{N-2}}{dt}=0$, and find

$$W_{N-2} = a_{N-2} W_{N-3} + b_{N-2} W_{N-4}$$

with the coefficients

$$a_{N-2} = -\frac{M_{N-2,N} a_N + M_{N-2,N-1} b_{N-1} + M_{N-2,N-3}}{M_{N-2,N} a_N + M_{N-2,N-1} a_{N-1} + M_{N-2,N} b_N + M_{N-2,N-2}},$$

and

$$b_{N-2} = -\frac{M_{N-2,N-4}}{M_{N-2,N} a_N + M_{N-2,N-1} a_{N-1} + M_{N-2,N} b_N + M_{N-2,N-2}}.$$

Continuing with this iteration, one finds in general

$$W_n = a_n W_{n-1} + b_n W_{n-2}.$$

with the coefficients

$$a_n = -\frac{M_{n,n+2} a_{n+2} + M_{n,n+1} b_{n+1} + M_{n,n-1}}{M_{n,n+2} a_{n+2} + M_{n,n+1} a_{n+1} + M_{n,n+2} b_{n+2} + M_{n,n}},$$

and

$$b_n = -\frac{M_{n,n-2}}{M_{n,n+2} a_{n+2} + M_{n,n+1} a_{n+1} + M_{n,n+2} b_{n+2} + M_{n,n}},$$

which is the form obtained for a_{N-2}, b_{N-2} if one replaces $N-2$ by n. One continues the iteration until one gets to $\frac{dW_1}{dt}=0$ with the solution $W_1=a_1 W_0$ because $b_1=0$. With $W_n=c_n W_0$, one finds $c_1=a_1$ and from

$$W_2 = c_2 W_0 = a_2 W_1 + b_1 W_0, \quad c_2 = a_2 c_1 + b_1,$$

or in general

$$W_n = c_n W_0 = a_n W_{n-1} + b_n W_{n-2}, \quad c_n = a_n c_{n-1} + b_n c_{n-2},$$

up until $n=N$. W_0 finally is obtained from the normalization

$$W_0 + W_1 + \ldots + W_{N-1} + W_N = 1,$$

from which one gets

$$W_0 = \frac{1}{1 + c_1 + c_2 + \ldots + c_N}.$$

With this relation all probabilities W_n are determined uniquely.

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