Fully analytical framework for non-equilibrium phase transition to Bose–Einstein condensate

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The theoretical description of non-equilibrium Bose–Einstein condensate (BEC) is one of the main challenges in modern statistical physics and kinetics. The non-equilibrium nature of BEC makes it impossible to employ the well-established formalism of statistical mechanics. We develop a framework for the analytical description of a non-equilibrium phase transition to BEC that, in contrast to previously developed approaches, takes into account the infinite number of the states. We consider the limit of fast thermalization and obtain an analytical expression for the full density matrix of a non-equilibrium BEC which also covers the equilibrium case. For the particular cases of 2D and 3D, we investigate the non-equilibrium formation of BEC by finding the temperature dependence of the ground state occupation and second-order coherence function. We show that for a given pumping rate, the macroscopic occupation of the ground state and buildup of coherence occur at different temperatures. Moreover, the buildup of coherence strongly depends on the pumping scheme. We also investigate the condensate linewidth and show that the Schawlow–Townes law holds for BEC in 3D and does not hold for BEC in 2D.

1 Introduction

Non-equilibrium Bose–Einstein condensates (BECs) working at room temperature [1, 2] are systems where open dissipative quantum-mechanical nature meets many-body collective dynamics. Besides its fundamental theoretical interest, BEC can be used in sub-picosecond switching at the fundamental quantum limit [3], all-optical manipulation, and other low-energy optoelectronic applications [4, 5]. Among the most promising systems for the realization of BEC are exciton-polaritons, the hybrid quasiparticles formed by the strong light–matter interaction of photons in a cavity and excitons, [1, 2, 5–17]. The advantage of the exciton-polaritons is their small effective mass and low density of states [9, 10, 14, 16], that provides a critical temperature of hundreds of kelvin [1, 6, 7].

Previous studies of BEC in these systems largely rely on the semiclassical Maxwell–Boltzmann equations describing the average population of exciton-polariton states [18–22], other mean-field theories [23–25], and an approach beyond mean-field theory [26] that takes into account the higher-order correlations. However, in such treatments, information about the correlations between different polariton states is lost. In many cases, the driven dissipative Gross–Pitaevskii equation with noise successfully describes the dynamics of BEC [27]. However, the Gross–Pitaevskii equation works well only when temperature is well below the condensation temperature [27]. This means that it cannot accurately describe the crossover and buildup of coherence.
In many BEC realizations, the thermalization rate of the polaritons is much faster that their dissipation rate and pumping rate. Namely, for long-lived polariton and photon systems, thermalization occurs at pumping rates significantly lower than the condensation threshold \([13, 28]\). The relatively high thermal energy in polariton systems at room temperature facilitates phonon-and vibron-assisted polariton relaxation, which, overall, results in efficient polariton thermalization. A fast thermalization in an organic polariton system has been revealed with an extreme rate on the order of 200 fs \([29, 30]\).

Recently it has been shown that significant advancement in the description of non-equilibrium BEC can be achieved in the case of fast thermalization of polaritons. For such systems the full density matrix of the BEC can be obtained analytically from the Lindblad master equation \([31]\). This is a powerful approach, because the complete information about the non-equilibrium BEC can be accessed through this density matrix both below and above the condensation threshold. However, this approach \([31]\) was developed only for countable sets of polariton states. This limits its ability to analyze the influence of the polaritons’ dispersion and dimension on the condensation threshold, buildup of coherence, long-range spatial correlations, and linewidth. Thus, a comprehensive analysis of BEC requires the extension of this approach \([31]\) to an infinite number of states.

In the present paper, we develop a formalism which extends the advantages of the exactly solvable Lindblad master equation for non-equilibrium BEC in the fast thermalization limit \([31]\) to continuous systems with an infinite number of states. We obtain the full density matrix of all the states, which enables finding correlations of any order. We show that for a non-equilibrium BEC in the fast thermalization limit, the condition for the macroscopic occupation of the ground state is the same as in the full thermal equilibrium case. We show that the buildup of coherence in a non-equilibrium BEC depends not only on the number of polaritons and the environmental temperature, but also on the pumping scheme. An increase in the pumping rate of the ground state lead to the macroscopic occupation of the ground state, but destroys the coherence of a non-equilibrium BEC. However, an increase in the pumping rate of the excited states results in both the macroscopic occupation of the ground state and the buildup of coherence. We also show that the linewidth of the condensate narrows with an increase of the macroscopic occupation of the ground state, but in 2D this narrowing does not follow the Schawlow–Townes equation.

2 DESCRIPTION OF THE MODEL

The strong light–matter interaction of a cavity electric field with the dipole moments of an optical transition of molecules gives rise to new eigenstates, namely, lower and upper polariton branches. Due to dissipation processes, it is necessary to compensate for the losses, which can be done by an external pump. Usually, an external pump excites polaritons in the upper polariton branch and excitons which are not coupled with the cavity modes \([2, 9]\). The polaritons of the upper branch and excitons \([2, 9]\) scatter the energy supplied by the pump into the lower polariton branch forming BEC at high enough excitation density. There are several mechanisms allowing such energy relaxation in polariton systems. Scattering by electrons \([32]\), high-energy optical phonons \([33]\), and vibrons in organic materials \([2, 34]\) are among them. Since non-equilibrium BEC occurs mostly in the lower polariton branch, below we consider the dynamics of the lower polariton branch only. The energy transfer from the upper polaritons or excitons to the lower polaritons will be described by an effective incoherent pumping. Such an approximation is reasonable for the majority of experimental realizations \([1, 7, 13]\).

We consider a continuous system of polariton states with the frequency \(\omega_k\) for a wave vector \(k\) in the general case of \(D\) dimensional space. We suppose that the polaritons with wave vector \(k\) of the lower polariton branch are described by bosonic creation \(\hat{a}_k\) and annihilation \(\hat{a}^\dagger_k\) operators \([35, 36]\) which obey the commutation relation \([\hat{a}_k, \hat{a}^\dagger_q] = \delta(k - q)\). In this case, the Hamiltonian of the polaritons takes the form

\[
\hat{H}_{LP} = \sum_k \omega_k \hat{a}^\dagger_k \hat{a}_k, \tag{1}
\]

where the state with \(k = 0\) is the ground state. Hereafter, we assume that \(\hbar = 1\).
We describe the dynamics of the polaritons in the lower polariton branch through the density matrix \( \dot{\rho} \). We consider the relaxation and pumping processes in the Born–Markov approximation [37]. In this approximation, the density matrix is governed by the master equation in the Lindblad form. The dissipation of the lower polaritons is described by the Lindblad superoperator [35]

\[
L_{\text{diss}}(\dot{\rho}) = \sum_k \gamma_k \left( \hat{a}_k \hat{\rho} \hat{a}_k^\dagger - \frac{1}{2} \hat{\rho} \hat{a}_k^\dagger \hat{a}_k - \frac{1}{2} \hat{a}_k^\dagger \hat{a}_k \hat{\rho} \right),
\]

where \( \gamma_k \) is the dissipation rate of the state with wave vector \( \mathbf{k} \). The effective incoherent pumping of the lower polaritons can be described by the Lindblad superoperator

\[
L_{\text{pump}}(\dot{\rho}) = \sum_k \kappa_k \left( \hat{a}_k \hat{\rho} \hat{a}_k^\dagger - \frac{1}{2} \hat{\rho} \hat{a}_k^\dagger \hat{a}_k - \frac{1}{2} \hat{a}_k^\dagger \hat{a}_k \hat{\rho} \right)
\]

where \( \kappa_k \) is the pumping rate of the state with wave vector \( \mathbf{k} \).

Thermalization of the lower polaritons may occur due to different physical processes depending on the system. For example, in organic polariton systems, thermalization occurs due to their nonlinear interaction with low frequency vibrations [25, 38–43]. For polariton states in inorganic semiconductors thermalization predominantly goes through interactions with acoustic phonons or free charges [35]. The thermalization of the polaritons can also occur due to polariton–polariton scattering [14, 44]. However, regardless of the mechanism, the thermalization can be described through the Lindblad superoperator [35, 36].

\[
L_{\text{therm}}(\dot{\rho}) = \sum_k \sum_{\mathbf{q} \neq \mathbf{k}} \Gamma_{\mathbf{kq}} x \left( \hat{a}_k \hat{\rho} \hat{a}_q^\dagger \hat{a}_q - \frac{1}{2} \hat{\rho} \hat{a}_q^\dagger \hat{a}_k \hat{a}_q^\dagger \hat{a}_q - \frac{1}{2} \hat{a}_q \hat{a}_k^\dagger \hat{a}_q \hat{\rho} \right),
\]

where \( \Gamma_{\mathbf{kq}} \) is the transition rate from the state with the wave vector \( \mathbf{k} \) to the state with the wave vector \( \mathbf{q} \). The thermalization rates \( \Gamma_{\mathbf{kq}} \) obey the Kubo–Martin–Schwinger relation \( \Gamma_{\mathbf{kq}} / \Gamma_{\mathbf{qk}} = \exp \left( (\omega_k - \omega_q) / T \right) \), where \( T \) is the temperature of intermolecular vibrations of the organic dyes or the temperature of the phonons in the semiconductors [45]. Hereafter, we assume the Boltzmann constant to be equal to unity.

Thus, the complete master equation for the density matrix of the polaritons of the lower polariton branch \( \dot{\rho} \) has the form

\[
\frac{d\rho(t)}{dt} = -i \left[ \rho(t), \hat{H}_{\text{LP}} \right] + L_{\text{diss}}(\dot{\rho}(t)) + L_{\text{pump}}(\dot{\rho}(t)) + L_{\text{therm}}(\dot{\rho}(t)).
\]

### 3 DENSITY MATRIX IN THE FAST THERMALIZATION LIMIT

Further, we assume that the thermalization is the fastest process in the system, i.e., thermalization rates are much larger than the dissipation and pumping rates, \( \Gamma_{\mathbf{0k}} (1 + \langle \hat{\alpha}_0^\dagger \hat{\alpha}_0 \rangle) \gg \kappa_k, \gamma_k \). In such a case, at times \( t \ll \gamma_k^{-1}, \kappa_k^{-1} \) only thermalization affects the system dynamics. As a result, the density matrix obeys the approximate differential equation

\[
\frac{d\rho(t)}{dt} = -i \left[ \rho(t), \hat{H}_{\text{LP}} \right] + L_{\text{therm}}(\dot{\rho}(t)).
\]

After the time \( \Gamma_{\mathbf{0k}}^{-1} (1 + \langle \hat{\alpha}_0^\dagger \hat{\alpha}_0 \rangle)^{-1} \ll t \ll \gamma_k^{-1}, \kappa_k^{-1} \) the system reaches its quasistationary state determined by

\[
- i \left[ \rho(t), \hat{H}_{\text{LP}} \right] + L_{\text{therm}}(\dot{\rho}(t)) = 0.
\]

The relaxation operator \( L_{\text{therm}}(\dot{\rho}) \), defined by the expression (4), conserves the total number of lower polaritons [35]. Thus, in the limit \( \kappa_k / \Gamma_{\mathbf{0k}} (1 + \langle \hat{\alpha}_0^\dagger \hat{\alpha}_0 \rangle) \rightarrow 0 \) and \( \gamma_k / \Gamma_{\mathbf{0k}} (1 + \langle \hat{\alpha}_0^\dagger \hat{\alpha}_0 \rangle) \rightarrow 0 \) the system reaches a quasistationary state governed by Eq. (7) at instantaneous time conserving the total number of polaritons. Applying the theory developed in [46], we can obtain the general form of the density matrix of this quasi-stationary state. Because the relaxation operator \( L_{\text{therm}}(\dot{\rho}) \) conserves the total number of lower polaritons [35], the operator of total number of lower polaritons \( \sum_k \hat{\alpha}_k^\dagger \hat{\alpha}_k \) is an integral of motion for the thermalization process. According to [46], the presence of this integral of motion implies that the system has invariant subspaces with the total number of polaritons equal to \( \sum_k n_k = N \). Being in an invariant subspace
at the initial moment in time, the system stays in this invariant subspace in the subsequent moments. In each invariant subspace with \(N\) polaritons, the Gibbs distribution with a temperature \(T\) is established [46].

The most general form of density matrix satisfying Eq. (7) is the weighted sum of thermalized distributions in each of the invariant subspaces:

\[
\hat{\rho}(t) = \sum_{N=0}^{\infty} P_N(t) \hat{\rho}_N, \tag{8}
\]

where the summation goes over the total number of polaritons. Here \(P_N(t)\) is the probability of the system to be in a state with exactly \(N\) polaritons total. These probabilities depend on the time due to the dissipation and pumping processes. \(\hat{\rho}_N\) is the thermalized density matrix of the states with exactly \(N\) polaritons, which forms an invariant subspace (see [31]). This means that \(\hat{\rho}_N\) can be represented as

\[
\hat{\rho}_N = \frac{1}{Z_N} \sum_{\text{config}: \sum_k n_k=N} e^{-\sum_k n_k(\omega_k - \omega_0)/T} \hat{R}_{\text{config}}, \tag{9}
\]

where \(Z_N\) is the partition function of the states with exactly \(N\) polaritons, and \(\hat{R}_{\text{config}}\) is a diagonal density matrix corresponding to the configuration of the polaritons \(\{n_k\}\). This density matrix obeys \(\hat{a}_k^\dagger \hat{a}_k \hat{R}_{\text{config}} = n_k \hat{R}_{\text{config}}, \sum n_k = N\).

The partition function \(Z_N\) is defined by the normalization condition \(\text{Tr}(\hat{R}_{\text{config}}) = 1\), thus

\[
Z_N = \sum_{\text{config}: \sum_k n_k=N} e^{-\sum_k n_k(\omega_k - \omega_0)/T}. \tag{10}
\]

When there are no polaritons in the system, Eq. (9) implies \(\hat{\rho}_{N=0} = Z_0^{-1}\langle 0|\langle 0|\). Therefore,

\[
Z_0 = 1. \tag{11}
\]

The density matrix (8) completely characterizes the condensate. In particular, it allows calculating the averages \(\langle \hat{n}_{k_1}^{m_1} \ldots \hat{n}_{k_M}^{m_M} \rangle:\)

\[
\langle \hat{n}_{k_1}^{m_1} \ldots \hat{n}_{k_M}^{m_M} \rangle = (-T) \sum_{j=1}^{M} \sum_{N=0}^{\infty} \frac{P_N(t) \partial^{m_1}_{k_1} \ldots \partial^{m_M}_{k_M} Z_N}{Z_N} \frac{1}{\omega_k^{m_1} \ldots \omega_k^{m_M}} \tag{12}
\]

The derivative of \(Z_N\) with respect to \(\omega_k\) is (see the Appendix)

\[
\frac{\partial Z_N}{\partial \omega_k} = \frac{1}{T} \left[Z_N - \sum_{n=0}^{N} Z_{N-n} e^{-n(\omega_k - \omega_0)/T}\right]. \tag{13}
\]

Note that \(\frac{\partial Z_N}{\partial \omega_0}\) must be calculated as \(\lim_{k \to 0} \frac{\partial Z_N}{\partial \omega_k}\).

4. PARTITION FUNCTION \(Z_N\)

The aim of paper work is to study the process of formation of BEC and its properties in the continuous limit. For this purpose, we consider the continuous limit of the partition function (10) and replace the sum over wave vectors \(k\) by the integral over frequencies \(\omega\):

\[
\sum_k \ldots = \int d\omega \left(1 \cdot \delta(\omega - \omega_0) + \nu(\omega)\right) \ldots \tag{14}
\]

The density of states \(\nu(\omega)\) is

\[
\nu(\omega) = \frac{V}{(2\pi)^D} \frac{\partial D}{\partial \omega_k} \omega_k = \omega \tag{15}
\]

where \(V\) is the volume in 3D or the area in 2D, occupied by polaritons. The transition to the continuous system is accurate if the differences between eigenfrequencies are much less than \(T\) [47]. Note that the delta function with prefactor unity in the right-hand side of Eq. (14) suggests that at \(k = 0\) there is only one state. Such a separation of the state \(k = 0\) is a standard step in the description of continuous systems [48]. Below we consider the quadratic dispersion, \(\omega_k = \omega_0 + \alpha k^2\). This dispersion is characteristic for polaritons of the lower branch in the vicinity of \(k = 0\) and the most frequent case in the experimental realizations of BEC.

The definition of the partition function (10) cannot be used directly to calculate \(Z_N\) in the continuous limit. The fastest way to calculate \(Z_N\) is to use a recurrence relation. The recurrence relation follows from the fact that the density matrix (9) corresponds to exactly \(N\) polaritons in all the states. Thus

\[
N = \sum_k \langle \hat{n}_k \rangle_N, \tag{16}
\]

where \(\langle \hat{n}_k \rangle_N\) stands for \(\text{tr}(\hat{n}_k \hat{\rho}_N)\). According to Eqs. (12) and (13),

\[
\sum_k \langle \hat{n}_k \rangle_N = \sum_{n=1}^{N} \frac{Z_{N-n}}{Z_N} \sum_k e^{-n(\omega_k - \omega_0)/T}. \tag{17}
\]
After integration over $k$ we obtain the recurrence relations

$$Z_N = \frac{1}{N} \sum_{n=0}^{N-1} Z_n \left(1 + \frac{G_{2D}}{N - n}\right)$$  \hspace{1cm} (18)

with $G_{2D} = VT/4\pi\alpha$ in 2D and

$$Z_N = \frac{1}{N} \sum_{n=0}^{N-1} Z_n \left(1 + \frac{G_{3D}}{(N - n)^{3/2}}\right)$$  \hspace{1cm} (19)

with $G_{3D} = V(T/4\pi\alpha)^{3/2}$ in 3D. In both cases the initial value is $Z_0 = 1$ (see Eq. (11)). The physical meaning of $G_{2D}$ and $G_{3D}$ is the number of states in the energy range $T$ above $k = 0$ in the corresponding dimensions.

Exact expressions for $Z_N$ can be obtained through its generating function (see the Appendix). However, recall that the optimal strategy to calculate $Z_N$ is to use the recurrence relations (18) and (19).

5 CONDENSATION OF EXACTLY $N$ POLARITONS

In Section 3 we showed that in the fast thermalization limit the invariant subspaces play a substantial role in the dynamics of BEC. In this section, we explore the formation of BEC in these invariant subspaces. This means that in Eq. (8) only one probability $P_N$ is non-zero, and it is equal to 1. Therefore, the system is described by the density matrix (9) and has exactly $N$ polaritons in all the states.

From Eq. (12) it follows that the average number of polaritons in the state with the wave vector $k$ can be calculated as

$$\langle \hat{n}_k \rangle_N = \sum_{n=1}^{N} \frac{Z_N - n}{Z_N} e^{-n(\omega_k - \omega_0)/T}.$$  \hspace{1cm} (20)

The subscript $N$ refers to the total number of polaritons in all the states. This expression is the same both in 2D and in 3D.

Further consideration substantially depends on the condition in which we consider Bose–Einstein condensation.

5.1 Infinite particle number and volume, finite density

The standard approach [48] is to consider the limiting case of infinite volume, $V \to \infty$, and infinite number of particles, $N \to \infty$, keeping the particle density constant, $N/V \to \text{const}$. It is known, that in such a limit, BEC forms in the 3D case and does not form in the 2D case.

Our theory reproduces these well-known results. Indeed, in the limit $N \to \infty$, from Eq. (20) we obtain the fraction of polaritons in the ground state (see the Appendix)

$$\frac{\langle \hat{n}_k = 0 \rangle_N}{N} \approx 1 - \left(\frac{T}{4\pi\alpha}\right) \left(\frac{V}{N}\right) \ln(N)$$  \hspace{1cm} (21)

in 2D and

$$\frac{\langle \hat{n}_k = 0 \rangle_N}{N} \approx 1 - \left(\frac{T}{4\pi\alpha}\right)^{3/2} \left(\frac{V}{N}\right) \zeta(3/2)$$  \hspace{1cm} (22)

in 3D. Here $\zeta(3/2)$ is the zeta function evaluated at $3/2$. One can see from Eqs. (21) and (22) that in the limit $V \to \infty$, $N/V \to \text{const}$, in the 3D case the fraction of the particles in the ground state becomes macroscopic (compared to unity). In contrast, in the 2D case, in the limit $N, V \to \infty$, $N/V \to \text{const}$, at any finite temperature, the number of particles in the ground state is infinitesimally small due to the factor $\ln(N)$.

5.2 Finite volume, particle number and density

Due to the external pumping, one can consider the case when the volume is finite and fixed and the number of particles may vary. In such a case, Bose–Einstein condensation occurs both in 2D and in 3D at low temperatures (Fig. 1). Indeed, if in Eqs. (21) and (22) we fix the volume and let the particle number go to infinity, the particle number in the ground state will tend to unity as $1 - \ln(N)/N$ in 2D and as $1 - 1/N$ in 3D. In many experiments, it is the external pump which is the control on the total number of particles while the volume is constant. Therefore, we have a situation where the volume is finite while the particle number and the density grow. For this reason, non-equilibrium BEC is possible both in 2D and in 3D. In what follows, we explore in more detail the case of finite and fixed volume.

Entropy. The density matrix (9) allows obtaining the entropy, $S_N = -\text{tr}(\hat{\rho}_N \ln \hat{\rho}_N)$, in 2D

$$S_N = \ln Z_N + G_{2D} \sum_{n=1}^{N} \frac{Z_N - n}{Z_N} \frac{1}{n^2}$$  \hspace{1cm} (23)
Figure 1: The fraction of the polaritons in the excited state in 2D (a) and in 3D (c), given that there are exactly \( N \) polaritons in the system. The entropy per polariton in 2D (b) and in 3D (d), given that there are exactly \( N \) polaritons in the system. The temperature is in the arbitrary units, \( T = 1 \) corresponds to \( G_{2D} = 1 \) in 2D and \( G_{3D} = 1 \) in 3D.

and in 3D

\[
S_N = \ln Z_N + \frac{3}{2} G_{3D} \sum_{n=1}^{N} \frac{Z_{N-n}}{Z_N} \frac{1}{n^{5/2}}.
\]

The entropy per polariton decreases (Fig. 1) as the total number of polaritons grows. This is because an increase in the number of polaritons leads to an increase in the ratio of polaritons that are in the ground state.

**First- and second-order coherence function.** The first-order coherence function is defined as [21, 49]

\[
g_{N}^{(1)}(r) = \frac{\langle \hat{\psi}^\dagger(0)\hat{\psi}(r) \rangle_N}{\sqrt{\langle \hat{\psi}^\dagger(0)\hat{\psi}(0) \rangle_N} \sqrt{\langle \hat{\psi}^\dagger(r)\hat{\psi}(r) \rangle_N}},
\]

where \( \hat{\psi}(r) \) is a wave function with the following plane-wave expansion:

\[
\hat{\psi}(r) = \frac{1}{\sqrt{V}} \sum_{k} \hat{a}_k e^{ikr}.
\]

From Eq. (9) it follows that \( \langle \hat{a}_k^\dagger \hat{a}_{k'} \rangle = \langle \hat{n}_k \rangle \delta_{kk'} \).

Thus, we obtain

\[
g_{N}^{(1)}(r) = \frac{\langle \hat{n}_{k=0} \rangle_N}{N} +
G_{2D} \frac{N}{N} \sum_{n=1}^{N} \frac{Z_{N-n}}{Z_N} \frac{1}{n} \exp \left[ -\frac{1}{n} \left( \frac{G_{2D} r}{2\sqrt{\alpha}} \right)^2 \right]
\]

in 2D and

\[
g_{N}^{(1)}(r) = \frac{\langle \hat{n}_{k=0} \rangle_N}{N} +
G_{3D} \frac{N}{N} \sum_{n=1}^{N} \frac{Z_{N-n}}{Z_N} \frac{1}{n^{3/2}} \exp \left[ -\frac{1}{n} \left( \frac{G_{3D} r}{2\sqrt{\alpha}} \right)^2 \right],
\]

in 3D. The definition (25) implies that \( g_{N}^{(1)}(0) = 1 \). However, \( g_{N}^{(1)}(r) = 0 \) as \( r \to +\infty \) only below the condensation threshold (see Eqs. (27) and (28)). This means that a non-equilibrium transition to BEC is always accompanied by the buildup of long-range spatial correlations.

Using Eq. (12) we obtain the second-order coherence function of the ground state

\[
\langle g_{k=0}^{(2)} \rangle_N = \frac{\langle \hat{n}_{k=0} \hat{n}_{k=0} \rangle_N - \langle \hat{n}_{k=0} \rangle_N^2}{\langle \hat{n}_{k=0} \rangle_N^2},
\]

where

\[
\langle \hat{n}_{k=0} \hat{n}_{k=0} \rangle_N = \sum_{n=1}^{N} \frac{Z_{N-n}}{Z_N} (2n - 1).
\]

The subscript \( N \) indicates that there are exactly \( N \) polaritons total in the system. For \( N \gg 1 \), a decrease in temperature leads to a drop in the second-order correlation function from \( \approx 2 \) to
Thus, for a fixed total number of polaritons, the formation Bose–Einstein condensation is accompanied by a buildup of coherence (Fig. 2).

6 KINETICS OF THE CONDENSATION

In the previous section, we considered the BEC when there are exactly \( N \) polaritons in the system. In this section, we consider the kinetics of the condensation and take into account a dissipation and pumping that do not change in time. Generally, there are two conditions for the formation of condensate. The first condition is the microscopic occupation of the ground state. This condition means that \( \langle \hat{N} \rangle \) and \( T \) are in the condensation region in the \( \{N, T\} \) plane (see Fig. 1). The second condition is the buildup of coherence in the ground state. This condition means that the second-order correlation function reaches 1. Below, we show that in non-equilibrium BEC in the fast thermalization limit these two conditions differ.

Due to dissipation and pumping processes, all the probabilities \( P_N \) differ from zero and may depend on time. To obtain \( P_N \) we substitute the thermalized density matrix (8) into Eq. (5). As a result, we obtain

\[
\frac{\partial P_0(t)}{\partial t} = \frac{d_0}{Z_1} P_1(t) - \frac{p_0}{Z_0} P_0(t),
\]

(31)

\[
\frac{\partial P_N(t)}{\partial t} = \frac{d_N}{Z_N+1} P_{N+1}(t) - \frac{d_{N-1} + p_N}{Z_N} P_N(t) + \frac{p_{N-1}}{Z_{N-1}} P_{N-1}(t)
\]

(32)

for \( N > 0 \). The coefficients \( p_N \) and \( d_N \) are defined by

\[
d_N = \sum_{n=0}^{N} Z_{N-n} \sum_{k} (\gamma_k + \kappa_k) e^{-(n+1)(\omega_k - \omega_0)/T},
\]

(33)

\[
p_N = \sum_{n=0}^{N} Z_{N-n} \sum_{k} \kappa_k e^{-n(\omega_k - \omega_0)/T}.
\]

(34)

The stationary solution of Eqs. (31)–(32) can be obtained analytically in the form of the recurrence relation

\[
P_1 = \frac{Z_1}{Z_0} \frac{p_0}{d_0} P_0,
\]

(35)

\[
P_{N+1} = \frac{Z_{N+1} d_{N-1} + p_N}{Z_N} P_N - \frac{Z_{N+1} p_{N-1}}{Z_{N-1}} \frac{d_N}{d_{N-1}} P_{N-1}
\]

(36)
for \( N \geq 1 \). \( P_0 \) in (35) is defined by the normalization condition \( \sum_{N=0}^{+\infty} P_N = 1 \).

From Eq. (8) it follows, that both the particle number in the ground state \( \langle \hat{n}_{k=0} \rangle \) and the entropy \( S \) can be expressed through the probabilities \( P_N \):

\[
\langle \hat{n}_{k=0} \rangle = \sum_{N=0}^{+\infty} P_N \langle \hat{n}_{k=0} \rangle_N, \quad (37)
\]

\[
S = \sum_{N=0}^{+\infty} P_N S_N - \sum_{N=0}^{+\infty} P_N \ln P_N, \quad (38)
\]

where \( \langle \hat{n}_{k=0} \rangle \) is defined by Eq. (20) and \( S_N \) is defined by Eq. (23) in 2D and by Eq. (24) in 3D. This means that the total average number of polaritons is the sum over the invariant subspaces weighted with the probabilities of the system being in the corresponding subspace.

Unlike the average number of polaritons in the ground state, \( g^{(1)}(\mathbf{r}) \) and \( g^{(2)}_{k=0}(0) \) are not the weighted sum of \( g^{(1)}_N(\mathbf{r}) \) and \( (g^{(2)}_{k=0}(0))_N \) over \( V \), but instead polaritons in each of the invariant subspace are in the Fock state. Nevertheless, this does not imply a buildup of the coherence because the dissipation and the pumping processes should be taken into account.

The coherence of the polaritons in the ground state is completely determined by the reduced density matrix of the ground state, \( \hat{\rho}_{k=0} \). At \( T = 0 \) this reduced density matrix follows straightforwardly from Eq. (8)

\[
\hat{\rho}_{k=0} = \sum_{n=0}^{+\infty} \vert n \rangle \langle n \vert P_n. \quad (41)
\]

The stationary solution (35)–(36) for probabilities \( P_N \) in this case reduces to

\[
P_N = \frac{1}{N!} \left( \frac{\kappa_{k=0}}{\gamma_{k=0} + \kappa_{k=0}} \right)^N \left( 1 - \frac{\kappa_{k=0}}{\gamma_{k=0} + \kappa_{k=0}} \right)^{-(1+\kappa_1/\kappa_{k=0})} f_N \left( 1 + \frac{\kappa_1}{\kappa_{k=0}} \right), \quad (42)
\]

where \( \gamma_{k=0} \) and \( \kappa_{k=0} \) are the dissipation and the pumping rates for \( k = 0 \), and \( \kappa_1 \) is the total pumping rate of the all excited states

\[
\kappa_1 = \sum_{k \neq 0} \kappa_k. \quad (43)
\]

The function \( f_N \) is determined by

\[
f_0(x) = 1, \quad (44)
\]

\[
f_N(x) = x(x+1)...(x+N-1) \quad (45)
\]

for \( N > 0 \).
ized to the ground state and long-range spatial correlations are established. But different pumping schemes result in different statistics of the condensate. When both the ground and excited states are pumped, $g_k^{(2)}(0) = 1$ and then find the average where at the initial moment in time (35)

$$\langle \hat{a}_k^\dagger(t)\hat{a}_k(t+\tau) \rangle \approx \hat{a}_k L_{\text{diss}}(\hat{\rho}(\tau))\hat{a}_k^\dagger + L_{\text{therm}}(\hat{\rho}(\tau)) \hat{a}_k^\dagger. \tag{53}$$

7 SPECTRAL WIDTH OF THE CONDENSATE

In this section, we explore the linewidth of the stationary BEC emission under constant pumping. The stationary emission spectrum, $I_k(\omega)$, of the polaritons with the wave vector $k$ is determined by the correlation function $\langle \hat{a}_k^\dagger(t)\hat{a}_k(t+\tau) \rangle$.\textsuperscript{37, 49}

$$I_k(\omega) = \lim_{t \to +\infty} \text{Re} \int_0^{+\infty} \langle \hat{a}_k^\dagger(t)\hat{a}_k(t+\tau) \rangle e^{-i\omega \tau} d\tau. \tag{50}$$

According to the quantum regression theorem\textsuperscript{37, 49} the correlation $\langle \hat{a}_k^\dagger(t)\hat{a}_k(t+\tau) \rangle$ obeys

$$\frac{d}{d\tau} \text{tr} \left( \hat{a}_k^\dagger \hat{\rho}(\tau) \hat{a}_k \right) = \text{tr} \left( \hat{a}_k L_{\text{pump}}(\hat{\rho}(\tau))\hat{a}_k^\dagger \right) \tag{51}$$

where at the initial moment in time $\hat{\rho}(0)$ is defined by the stationary solution (35)–(36). To obtain $\langle \hat{a}_k^\dagger(t)\hat{a}_k(t+\tau) \rangle$ we solve the Eq. (51) in the matrix form

$$\hat{a}_k^\dagger \frac{d\hat{\rho}(\tau)}{d\tau} \hat{a}_k = \hat{a}_k L_{\text{pump}}(\hat{\rho}(\tau))\hat{a}_k^\dagger \tag{52}$$

and then find the average $\langle \hat{a}_k^\dagger(t)\hat{a}_k(t+\tau) \rangle$.

In the fast thermalization limit, we may neglect $\hat{a}_k L_{\text{pump}}(\hat{\rho}(\tau))\hat{a}_k^\dagger$ and $\hat{a}_k L_{\text{diss}}(\hat{\rho}(\tau))\hat{a}_k^\dagger$ because these terms are small compared to $\hat{a}_k L_{\text{therm}}(\hat{\rho}(\tau))\hat{a}_k^\dagger$. As a result, Eq. (52) becomes

$$\hat{a}_k^\dagger \frac{d\hat{\rho}(\tau)}{d\tau} \hat{a}_k \approx \hat{a}_k L_{\text{therm}}(\hat{\rho}(\tau))\hat{a}_k^\dagger + L_{\text{therm}}'(\hat{\rho}(\tau)) \hat{a}_k^\dagger. \tag{53}$$

where

$$L_{\text{therm}}'(\hat{\rho}) = \hat{a}_k^\dagger \sum_{q\neq k} \Gamma_{kq} \hat{a}_q \hat{a}_q^\dagger \hat{a}_k \tag{54}$$

To solve Eq. (53), we assume that $L_{\text{therm}}$ in the right-hand side of Eq. (53) leads to a decay with the characteristic thermalization rate $\Gamma$. We also assume that $L_{\text{therm}}'$ leads to a decay with a rate much lower than $\Gamma$, but much higher than the dissipation rate and the pumping rate. After obtaining the solution of Eq. (53), we explicitly check these assumptions. Employing these assumptions, we search for a matrix $\rho(\tau)$ satisfying Eq. (53) in the form (8). As a result, we obtain

$$\langle \hat{a}_k^\dagger(t)\hat{a}_k(t+\tau) \rangle = \langle \hat{n}_k(t) \rangle_N e^{-\langle \Delta \omega_k \rangle N \tau^2} \tag{55}$$

as $t \to +\infty$, where

$$\langle \Delta \omega_k \rangle_N = \sum_{q \neq k} \Gamma_{kq} - \sum_{q \neq k} \frac{(\Gamma_{kq} - \Gamma_{kq} \langle \{1 + \hat{n}_q \} \hat{n}_q \rangle_N - 1)}{\langle 1 + \hat{n}_q \rangle_N - 1}. \tag{56}$$

From Eq. (55) it follows that the linewidth depends on the distribution of the stationary probabilities $P_N$ defined by Eqs. (35)–(36). The linewidth $\Delta \omega_k$ of the polaritons in the state $k$ can be estimated to be

$$\Delta \omega_k = \sum_{N=0}^{+\infty} P_N(0)(\Delta \omega_k)_N. \tag{57}$$

We consider in more detail the linewidth of the ground state, $(\Delta \omega_k)_N$. For simplicity we assume that the thermalization rates do not depend on the states, i.e., $\Gamma_{q0} = \Gamma$. Note that the thermalization rates obey the Kubo–Martin–Schwinger relation, $\Gamma_{0q} = \Gamma \exp(-\langle \omega_q - \omega_0 \rangle/T)$. From Eq. (12), (15) and (56) we obtain

$$(\Delta \omega_{k=0})_{N+1} = \Gamma \int_0^{+\infty} d\omega \nu(\omega + \omega_0) \sum_{n=0}^N Z_{N-n} e^{-(n+1)\omega/T} \frac{Z_n}{\sum_{n=0}^N Z_n}. \tag{58}$$

where $\nu(\omega)$ is the density of states defined by Eq. (15).
When $T$ is above the condensation temperature, the linewidth of the ground state emission is almost independent of the total number of polaritons (Fig. 2). But when $T$ is below the condensation temperature, an increase in $N$ leads to a narrowing of the linewidth (Fig. 2).

**Linewidth of the ground state in 2D.**

In 2D $(\Delta \omega_{k=0})_N$ is

$$
(\Delta \omega_{k=0})_{N+1} = \Gamma \frac{\sum_{n=0}^{N} Z_{N-n} (n+1)^{-1}}{\sum_{n=0}^{N} Z_n} G_{2D},
$$

(59)

where $G_{2D}$ is defined by Eq. (18). From Eq. (59) it follows that at low temperatures, when $G_{2D} \ll 1$, $(\Delta \omega_{k=0})_N \propto T$. Moreover, in this case $Z_n \approx 1$ for all $n$ and

$$
(\Delta \omega_{k=0})_{N+1} \approx \Gamma_1 (0) \frac{1}{N+1} \sum_{n=0}^{N} \frac{1}{n+1} \approx

\approx (\Delta \omega_{k=0})_{N=1} \frac{\ln N}{N},
$$

(60)

where $(\Delta \omega_{k=0})_{N=1}$ is the linewidth at $N = 1$. The last approximate equality in Eq. (60) holds for $N \gg 1$. Thus, in 2D the linewidth of the ground state deviates from the Schawlow–Townes law.

**Linewidth of the ground state in 3D.**

In 3D $\Gamma_N (0)$ is

$$
(\Delta \omega_{k=0})_{N+1} \approx \Gamma \frac{\sum_{n=0}^{N} Z_{N-n} (n+1)^{-3/2}}{\sum_{n=0}^{N} Z_n} G_{3D},
$$

(61)

where $G_{3D}$ is defined by Eq. (19). From (61) it follows that at low temperatures, when $G_{3D} \ll 1$, $(\Delta \omega_{k=0})_N \propto T^{3/2}$. Moreover, in this case $Z_n \approx 1$ for all $n$ and

$$
(\Delta \omega_{k=0})_{N+1} = \Gamma_1 (0) \frac{1}{N+1} \sum_{n=0}^{N} \frac{1}{(n+1)^{3/2}} \approx

\approx (\Delta \omega_{k=0})_{N=1} \frac{\zeta(3/2)}{N},
$$

(62)

where $(\Delta \omega_{k=0})_{N=1}$ is the linewidth at $N = 1$. The last approximate equality in (62) holds for $N \gg 1$. Thus, in 3D the linewidth follows the Schawlow–Townes law.

8 Conclusion

In conclusion, we developed a full analytical framework to describe Bose–Einstein condensates (BEC) in the fast thermalization limit. In this limit, we obtained an expression for the full density matrix of non-equilibrium BEC.

We showed that a macroscopic occupation of the ground state occurs under the same conditions for the full thermal equilibrium case as for the non-equilibrium case in the fast thermalization limit. The macroscopic occupation of the ground state is always accompanied by the formation of long-range first-order spatial correlations. Moreover, for a given system, only the temperature and the average number of the particles determine whether the ground state is macroscopically occupied. In contrast, the buildup of second-order coherence in BEC is not fully determined by the occupation of the ground state, but also depends on the pumping scheme. If excited states are pumped, then, above the condensation threshold, a coherence of the ground state is formed. If only the ground state is pumped, then the formation of ground state coherence does not occur even at zero temperature.

At fixed temperature, an increase in the number of polaritons leads to an increase in the total entropy, but the entropy per polariton decreases. This is because the polaritons in the ground state have low entropy and an increase in the number of the particles leads to an increase of the fraction of the particles in the ground state.

Above the condensation threshold, the linewidth of the ground state emission narrows. At low temperatures and large numbers of polaritons, this narrowing follows the Schawlow–Townes law in 3D, but in 2D the linewidth decreases more slowly than predicted by the Schawlow–Townes law. As the temperature decreases, the condensate line width decreases faster in 3D than in 2D.

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References

[1] Johannes D Plumhof, Thilo Stöferle, Lijian Mai, Ulrich Scherf, and Rainer F Mahrt. Room-temperature bose–einstein condensation of cavity exciton–polaritons in a polymer. *Nature materials*, 13(3):247–252, 2014. DOI: https://doi.org/10.1038/nmat3825.

[2] Anton V Zasedatelev, Anton V Baranikov, Darius Urbonas, Fabio Scafirimuto, Ulrich Scherf, Thilo Stöferle, Rainer F Mahrt, and Pavlos G Lagoudakis. A room-temperature organic polariton transistor. *Nature Photonics*, 13(6):378–383, 2019. DOI: https://doi.org/10.1038/s41566-019-0392-8.

[3] Anton V. Zasedatelev, Anton V. Baranikov, Denis Sannikov, Darius Urbonas, Fabio Scafirimuto, Vladislav Yu. Shishkov, Evgeny S. Andrianov, Yuriy E. Lozovik, Ulrich Scherf, Thilo Stöferle, Rainer F. Mahrt, and Pavlos G. Lagoudakis. Single-photon nonlinearity at room temperature. *Nature*, 597:493–497, 2021. DOI: https://doi.org/10.1038/s41586-021-03866-9.

[4] Daniele Sanvitto and Stéphane Kéna-Cohen. The road towards polaritonic devices. *Nature materials*, 15(10):1061–1073, 2016. DOI: https://doi.org/10.1038/nmat4668.

[5] Jonathan Keeling and Stéphane Kéna-Cohen. Bose–einstein condensation of exciton-polaritons in organic microcavities. *Annual Review of Physical Chemistry*, 71:435–459, 2020. DOI: https://doi.org/10.1146/annurev-physchem-010920-102509.

[6] Hui Deng, Gregor Weihs, David Snoke, Jacqueline Bloch, and Yoshihisa Yamamoto. Polariton lasing vs. photon lasing in a semiconductor microcavity. *Proceedings of the National Academy of Sciences*, 100(26):15318–15323, 2003. DOI: https://doi.org/10.1073/pnas.2634328100.

[7] Jacek Kasprzak, M Richard, S Kundermann, A Baas, P Jeambrun, JMJ Keeling, FM Marchetti, MH Szymańska, R André, JL Staehli, et al. Bose–einstein condensation of exciton polaritons. *Nature*, 443(7110):409–414, 2006. DOI: https://doi.org/10.1038/nature05131.

[8] Monique Combescot and Shiu-Yuan Shiau. *Excitons and Cooper pairs: two composite bosons in many-body physics*. Oxford University Press, 2015.

[9] Tim Byrnes, Na Young Kim, and Yoshihisa Yamamoto. Exciton–polariton condensates. *Nature Physics*, 10(11):803–813, 2014. DOI: https://doi.org/10.1038/nphys3143.

[10] Esther Wertz, Lydie Ferrier, DD Solnyshkov, Robert Johne, Daniele Sanvitto, Aristide Lemaître, Isabelle Sagnes, Roger Grousson, Alexey V Kavokin, Pascale Senellart, et al. Spontaneous formation and optical manipulation of extended polariton condensates. *Nature physics*, 6(11):860–864, 2010. DOI: https://doi.org/10.1038/nphys1750.

[11] R Balili, V Hartwell, D Snoke, L Pfeiffer, and K West. Bose-einstein condensation of microcavity polaritons in a trap. *Science*, 316(5827):1007–1010, 2007. DOI: https://doi.org/10.1126/science.1140990.

[12] E Estrecho, Tingge Gao, Nataliya Bobrovskaya, Michael D Fraser, M Steger, L Pfeiffer, K West, Timothy Chi Hin Liew, Michal Matuszewski, David W Snoke, et al. Single-shot condensation of exciton polaritons and the hole burning effect. *Nature communications*, 9(1):1–9, 2018. DOI: https://doi.org/10.1038/s41467-018-05349-4.

[13] Yongbao Sun, Patrick Wen, Yoseob Yoon, Gangqiang Liu, Mark Steger, Loren N Pfeiffer, Ken West, David W Snoke, and Keith A Nelson. Bose-einstein condensation of long-lifetime polaritons in thermal equilibrium. *Physical review letters*, 118(1):016602, 2017. DOI: https://doi.org/10.1103/PhysRevLett.118.016602.

[14] Hui Deng, Hartmut Haug, and Yoshihisa Yamamoto. Exciton-polariton bose-einstein condensation. *Reviews of Modern Physics*, 82(2):1489, 2010. DOI: https://doi.org/10.1103/RevModPhys.82.1489.

[15] M Klaas, E Schlottmann, H Flayac, FP Laussy, F Gericke, M Schmidt, M v Helversen, J Beyer, S Brodbeck, H Suchomel, et al. Photon-number-resolved measurement of an exciton-polariton condensate. *Physical
review letters, 121(4):047401, 2018. DOI: https://doi.org/10.1103/PhysRevLett.121.047401.

[16] A Imamoglu, RJ Ram, S Pau, Y Yamamoto, et al. Nonequilibrium condensates and lasers without inversion: Exciton-polariton lasers. Physical Review A, 53(6):4250, 1996. DOI: https://doi.org/10.1103/PhysRevA.53.4250.

[17] Mengjie Wei, Sai Kiran Rajendran, Hamid Ohadi, Laura Tropf, Malte C Gather, Graham A Turnbull, and Ifor DW Samuel. Low-threshold polariton lasing in a highly disordered conjugated polymer. Optica, 6(9):1124–1129, 2019. DOI: https://doi.org/10.1364/OPTICA.6.001124.

[18] Guillaume Malpuech, Aldo Di Carlo, Alexey Kavokin, Jeremy J Baumberg, Marian Zamfirescu, and Paolo Lugli. Room-temperature polariton lasers based on gan microcavities. Applied physics letters, 81(3):412–414, 2002. DOI: https://doi.org/10.1063/1.1494126.

[19] L Banyai and P Gartner. Real-time bose-einstein condensation in a finite volume with a discrete spectrum. Physical review letters, 88(21):210404, 2002. DOI: https://doi.org/10.1103/PhysRevLett.88.210404.

[20] Huy Thien Cao, TD Doan, DB Tran Thoai, and H Haug. Condensation kinetics of cavity polaritons interacting with a thermal phonon bath. Physical Review B, 69(24):245325, 2004. DOI: https://doi.org/10.1103/PhysRevB.69.245325.

[21] TD Doan, H Thien Cao, DB Tran Thoai, and H Haug. Coherence of condensed microcavity polaritons calculated within boltzmann-master equations. Physical Review B, 78(20):205306, 2008. DOI: https://doi.org/10.1103/PhysRevB.78.205306.

[22] F Tassone, C Piermarocchi, V Savona, A Quattropani, and P Schwendimann. Bottleneck effects in the relaxation and photoluminescence of microcavity polaritons. Physical Review B, 56(12):7554, 1997. DOI: https://doi.org/10.1103/PhysRevB.56.7554.

[23] Peter Kirton and Jonathan Keeling. Nonequilibrium model of photon condensation. Physical review letters, 111(10):100404, 2013. DOI: https://doi.org/10.1103/PhysRevLett.111.100404.

[24] Peter Kirton and Jonathan Keeling. Thermalization and breakdown of thermalization in photon condensates. Physical Review A, 91(3):033826, 2015. DOI: https://doi.org/10.1103/PhysRevA.91.033826.

[25] Artem Strashko, Peter Kirton, and Jonathan Keeling. Organic polariton lasing and the weak to strong coupling crossover. Physical Review Letters, 121(19):193601, 2018. DOI: https://doi.org/10.1103/PhysRevLett.121.193601.

[26] Kristin B Arnardottir, Antti J Moilanen, Artem Strashko, Päivi Törnä, and Jonathan Keeling. Multimode organic polariton lasing. Physical Review Letters, 125(23):233603, 2020. DOI: https://doi.org/10.1103/PhysRevLett.125.233603.

[27] Iacopo Carusotto and Cristiano Ciuti. Quantum fluids of light. Reviews of Modern Physics, 85(1):299, 2013. DOI: https://doi.org/10.1103/RevModPhys.85.299.

[28] Rafi Weill, Alexander Bekker, Boris Levit, and Baruch Fischer. Bose–einstein condensation of photons in an erbium–ytterbium co-doped fiber cavity. Nature communications, 10(1):1–6, 2019. DOI: https://doi.org/10.1038/s41467-019-08527-0.

[29] Tommi K Hakala, Antti J Moilanen, Aaro I Väkeväinen, Rui Guo, Jani-Petri Martikainen, Konstantinos S Daskalakis, Heikki T Rekola, Aleksi Julku, and Päivi Törnä. Bose–einstein condensation in a plasmonic lattice. Nature Physics, 14(7):739–744, 2018. DOI: https://doi.org/10.1038/s41567-018-0109-9.

[30] Aaro I Väkeväinen, Antti J Moilanen, Marek Nečada, Tommi K Hakala, Konstantinos S Daskalakis, and Päivi Törnä. Sub-picosecond thermalization dynamics in condensation of strongly coupled lattice plasmons. Nature communications, 11(1):1–12, 2020. DOI: https://doi.org/10.1038/s41467-020-16906-1.

[31] Vladislav Yu Shishkov, Evgeny S Andrianov, Anton V Zasedatelev, Pavlos G Lagoudakis, and Yurii E Lozovik. Exact analytical solution for density matrix of a non-equilibrium polariton bose-einstein condensate. arXiv preprint arXiv:2105.02940, 2021.

[32] PG Lagoudakis, MD Martin, JJ Baumberg, A Qarry, E Cohen, and LN Pfeiffer. Electron-polariton scattering in semiconductor microcavities. Physical
[30] M Litinskaya, P Reineker, and VM Agranovich. Fast polariton relaxation in strongly coupled organic microcavities. Journal of luminescence, 110(4):364–372, 2004. DOI: https://doi.org/10.1016/j.jlumin.2004.08.033.

[31] Leonard Mazza, L Fontanesi, and GC La Rocca. Organic-based microcavities with vibronic progressions: Photoluminescence. Physical Review B, 80(23):235314, 2009. DOI: https://doi.org/10.1103/PhysRevB.80.235314.

[32] Eric R Bittner and Carlos Silva. Estimating the conditions for polariton condensation in organic thin-film microcavities. The Journal of chemical physics, 136(3):034510, 2012. DOI: https://doi.org/10.1063/1.3678015.

[33] Justyna A Cwik, Sahinur Reja, Peter B Littlewood, and Jonathan Keeling. Polariton condensation with saturable molecules dressed by vibrational modes. EPL (Europhysics Letters), 105(4):47009, 2014. DOI: https://doi.org/10.1209/0295-5075/105/47009.

[34] N Somaschi, L Mouchliadis, D Coles, IE Perakis, DG Lidzey, PG Lagoudakis, and PG Savvidis. Ultrafast polariton population build-up mediated by molecular phonons in organic microcavities. Applied Physics Letters, 99(14):209, 2011. DOI: https://doi.org/10.1063/1.3645633.

[35] Mohammad Ramezani, Quynh Le-Van, Alexei Halpin, and Jaime Gómez Rivas. Nonlinear emission of molecular ensembles strongly coupled to plasmonic lattices with structural imperfections. Physical Review Letters, 121(24):243904, 2018. DOI: https://doi.org/10.1103/PhysRevLett.121.243904.

[36] PG Savvidis, JJ Baumberg, RM Stevenson, MS Skolnick, DM Whittaker, and JS Roberts. Angle-resonant stimulated polariton amplifier. Physical Review Letters, 84(7):1547, 2000. DOI: https://doi.org/10.1103/PhysRevLett.84.1547.

[37] Ronnie Kosloff. Quantum thermodynamics: A dynamical viewpoint. Entropy, 15(6):2100–2128, 2013. DOI: https://doi.org/10.3390/e15062100.

[38] V Yu Shishkov, ES Andrianov, AA Pukhov, AP Vinogradov, and AA Lisyansky. Zeroth law of thermodynamics for thermalized open quantum systems having constants of motion. Physical Review E, 98(2):022132, 2018. DOI: https://doi.org/10.1103/PhysRevE.98.022132.

[39] Oleg L Berman, Yuri E Lozovik, and David W Snoke. Theory of bose-einstein condensation and superfluidity of two-dimensional polaritons in an in-plane harmonic potential. Physical Review B, 77(15):155317, 2008. DOI: https://doi.org/10.1103/PhysRevB.77.155317.

[40] Lev Davidovich Landau and Evgenii Mikhailovich Lifshitz. Course of theoretical physics. Elsevier, 2013.

[41] Marlan O Scully and M Suhail Zubairy. Quantum optics, 1999.

[42] Louis Comtet. Advanced Combinatorics: The art of finite and infinite expansions. Springer Science & Business Media, 2012.
A Some properties of the partition function $Z_N$

In this appendix, we present some general properties of the partition function $Z_N$. To derive these properties we introduce the generating function of $Z_N$: $\sum_{N=0}^{+\infty} x^N Z_N$, $0 < x < 1$. Eq. (10) allows us to represent the generating function in different forms

$$\sum_{N=0}^{+\infty} x^N Z_N = \sum_{N=0}^{+\infty} x^N \sum_{\text{config}} \sum_{n_k=N}^\infty \sum_{n_k=0}^\infty e^{-\sum_k \frac{n_k}{T} (\omega_k - \omega_0)} = \prod_k \sum_{n_k=0}^{+\infty} e^{-n_k (\omega_k - \omega_0) / T - \ln x} = \prod_k \frac{1}{1 - x e^{-\omega_k / T}} = \exp \left\{ - \sum_k \ln \left[ 1 - x e^{-\omega_k / T} \right] \right\}. \tag{63}$$

The last two forms are especially useful for the considerations below.

A.1 Proof of Eq. (12)
From Eq. (63) it follows that

$$\sum_{N=0}^{+\infty} x^N \frac{\partial Z_N}{\partial \omega_q} = -\frac{1}{T} x e^{-\omega_q - \omega_0} \prod_k \frac{1}{1 - x e^{-\omega_k / T}} = -\frac{1}{T} x e^{-\omega_q - \omega_0} \sum_{N=0}^{+\infty} x^N Z_N = \sum_{N=0}^{+\infty} x^N \frac{1}{T} \left[ Z_N - \sum_{n=0}^{N} Z_{N-n} e^{n (\omega_q - \omega_0) / T} \right]. \tag{64}$$

Eq. (12) directly follows from this equation.

A.2 Monotonicity of $Z_N$
To prove the monotonicity of $Z_N$ we use Eq. (63) and consider the expression

$$\sum_{N=0}^{+\infty} x^N Z_N = \frac{1}{1-x} \sum_{N=0}^{+\infty} x^N F_N, \quad \tag{65}$$

where we introduced

$$\sum_{N=0}^{+\infty} x^N F_N = \prod_{k \neq 0} \frac{1}{1 - x e^{-\omega_k / T}}. \tag{66}$$

Although finding $F_N$ is hard in the general case, an important property of it follows directly from Eq. (66), namely $F_N > 0$. Moreover, from Eq. (65) it follows that $Z_N = \sum_{n=0}^{N} F_n$. Thus, $Z_N$ rises monotonously with $N$.

A.3 The limit of $Z_N$ as $N \to +\infty$
Below we assume that the limit of $Z_N$ as $N \to +\infty$ exists. In this case, we introduce the notation $Z_\infty = \lim_{N \to +\infty} Z_N$

$$Z_\infty = \lim_{N \to +\infty} \left[ \sum_{n=1}^{N} (Z_n - Z_{n-1}) + Z_0 \right] = \lim_{x \to 1-0} \lim_{N \to +\infty} \left[ \sum_{n=1}^{N} x^n (Z_n - Z_{n-1}) + x Z_0 \right] = \lim_{x \to 1-0} \lim_{N \to +\infty} \left[ \sum_{n=0}^{N-1} x^n Z_n - x \sum_{n=0}^{N-1} x^n Z_n \right] = \lim_{x \to 1-0} (1-x) \sum_{N=0}^{+\infty} x^N Z_N. \tag{67}$$

Thus, we express $Z_\infty$ through the generating function of $Z_N$. 

\[14\]
A.4 An exact expression for \( Z_N \) in 2D and in 3D for quadratic dispersion

To find an exact expression for \( Z_N \) we use its generating function (see Eq. (63))

\[
\sum_{N=0}^{+\infty} x^N Z_N = \exp \left\{- \sum_k \ln \left[ 1 - xe^{-(\omega_k - \omega_0)/T} \right] \right\}
\]

(68)

Below we consider the quadratic dispersion, \( \omega_k = \omega_0 + \alpha k^2 \), in cases of 2D and 3D.

Polaritons in 2D. For polaritons in 2D the density of states is \( \nu(\omega) = V/4\pi\alpha \). In this case, Eq. (68) takes the form

\[
\sum_{N=0}^{+\infty} x^N Z_N = \exp \left\{ G_{2D} \left[ \ln_2(x) \right] \right\}
\]

(69)

where \( G_{2D} = VT/4\pi\alpha \) is the number of states in the energy range \( T \) above \( k = 0 \) and \( \ln_2(x) \) is the polylogarithm of order 2.

Using the expansion of the exponent in the series of Bell polynomials [50], the expansion of \((1-x)^{-1}\) in a Taylor series, and equating the coefficients on the left and right sides of Eq. (69), we obtain

\[
Z_N = \sum_{n=0}^{N} \frac{1}{n!} B_n \left( \frac{1!}{1^2} G_{2D}, \frac{2!}{2^2} G_{2D}, \ldots, \frac{n!}{n^2} G_{2D} \right),
\]

(70)

where \( B_n \) is the \( n \)-th complete exponential Bell polynomial [50] and \( B_0 = 1 \). Thus, \( Z_N \) in 2D is fully determined by a dimensionless parameter \( G_{2D} \) and rises monotonically from \( Z_0 = 1 \) to \( Z_{\infty} = \exp(G_{2D}\zeta(2)) \), where \( \zeta(2) \) is the value of the zeta function at 2.

Polaritons in 3D. For polaritons in 3D case the density of states is \( \nu(\omega) = (V/4\pi^2\alpha)^{3/2} \sqrt{(\omega - \omega_0)/\alpha} \). In this case, Eq. (68) takes the form

\[
\sum_{N=0}^{+\infty} x^N Z_N = \exp \left\{ G_{3D} \left[ \ln_{5/2}(x) \right] \right\}
\]

(71)

where \( G_{3D} = V(T/4\pi\alpha)^{3/2} \) is the number of states in the energy range \( T \) above \( k = 0 \) and \( \ln_{5/2}(x) \) is the polylogarithm of the order \( 5/2 \).

From Eq. (71) it follows that

\[
Z_N = \sum_{n=0}^{N} \frac{1}{n!} B_n \left( \frac{1!}{1^{5/2}} G_{3D}, \frac{2!}{2^{5/2}} G_{3D}, \ldots, \frac{n!}{n^{5/2}} G_{3D} \right),
\]

(72)

Thus, \( Z_N \) in 3D is fully determined by the dimensionless parameter \( G_{3D} \) and rises monotonically from \( Z_0 = 1 \) to \( Z_{\infty} = \exp(G_{3D}\zeta(5/2)) \), where \( \zeta(5/2) \) is the value of the zeta function at \( 5/2 \).

The calculation \( Z_N \). We note that the direct application of Eq. (70) and (72) is not the fastest way to calculate \( Z_N \). This is due to the great computational difficulty of the complete Bell functions for \( N \gg 1 \). The optimal strategy to obtain \( Z_N \) is to use the corresponding recurrence relation, discussed in the main text.

B Proof of Eqs. (21) and (22)

According to Eq. (20), when \( N \gg 1 \)

\[
\frac{N - \langle \hat{n}_{k=0} \rangle}{N} = \frac{\sum_{n=0}^{N-1} (Z_N - Z_n)}{NZ_N} \approx \frac{\sum_{n=0}^{N-1} (Z_\infty - Z_n)}{NZ_\infty}.
\]

(73)
We consider \( \sum_{n=0}^{N-1} (Z_{\infty} - Z_n) \) in more detail.

In 2D we have

\[
\lim_{N \to +\infty} N^{-1} \sum_{n=0}^{N-1} (Z_{\infty} - Z_n) = \lim_{N \to +\infty} \lim_{x \to 1-0} \sum_{n=0}^{N-1} (Z_{\infty} - Z_n)x^n = \\
= \lim_{x \to 1-0} \frac{\exp(G_{2D} \operatorname{Ln}_2(1)) - \exp(G_{2D} \operatorname{Ln}_2(x))}{1-x} = Z_\infty G_{2D} \lim_{x \to 1-0} \operatorname{Ln}_1(x) = Z_\infty G_{2D} \lim_{N \to +\infty} \ln(N). \tag{74}
\]

Therefore, when \( N \gg 1 \)

\[
\frac{N - \langle \hat{n}_{k=0} \rangle}{N} \approx \frac{G_{2D}}{N} \ln N \tag{75}
\]

Eq. (21) directly follows from Eq. (75).

In 3D we have

\[
\lim_{N \to +\infty} N^{-1} \sum_{n=0}^{N-1} (Z_{\infty} - Z_n) = \lim_{N \to +\infty} \lim_{x \to 1-0} \sum_{n=0}^{N-1} (Z_{\infty} - Z_n)x^n = \\
= \lim_{x \to 1-0} \frac{\exp(G_{3D} \operatorname{Ln}_{5/2}(1)) - \exp(G_{3D} \operatorname{Ln}_{5/2}(x))}{1-x} = Z_\infty G_{3D} \lim_{x \to 1-0} \operatorname{Ln}_{3/2}(x) = Z_\infty G_{3D} \zeta(3/2). \tag{76}
\]

Therefore, when \( N \gg 1 \)

\[
\frac{N - \langle \hat{n}_{k=0} \rangle}{N} \approx \frac{G_{3D}}{N} \zeta(3/2). \tag{77}
\]

Eq. (22) directly follows from Eq. (77).