We develop a kinetic theory of microcavity polaritons in presence of both Coulomb and polariton-phonon interaction, obeying particle number conservation. We study the growth of a macroscopic population of condensed particles in the lowest polariton state, under steady-state incoherent excitation of higher energy states. The collective excitation spectrum, resulting from the Coulomb Hamiltonian treated within the Hartree-Fock-Bogolubov framework, strongly influences the polariton condensation kinetics. In particular, for values of the excitation intensity above the condensation threshold, scattering from the condensate into the collective excitation modes results in strong quantum fluctuations that deplete the condensate. A numerical evaluation based on a few-level scheme shows that the condensate fraction is expected to be lower than 1 even far above threshold. With increasing system size, the role of the polariton quantum fluctuations becomes dominant, eventually preventing condensation to occur for system size larger than 100 μm.

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I. INTRODUCTION

Quantum fluids are the most remarkable manifestation of quantum mechanics at the macroscopic scale. Superconductivity, superfluidity, and more recently Bose-Einstein condensation (BEC) of diluted atoms are all examples of a system in which many particles share the same quantum mechanical wave function. A long sought and never observed quantum fluid is the BEC of excitons in semiconductors. Presently, it is not well understood why excitonic BEC eludes experimental observation. Presumably however, three factors are believed to play against BEC. First the structural disorder, which induces a fragmentation of the condensate, effectively increasing the condensation critical density as pointed out by Nozieres. Second, the high rate of exciton-exciton Coulomb scattering, expected to cause a strong dephasing of the condensate already at moderate density, which is not predicted by the standard mean-field approach to BEC. Third, the strong composite boson nature of excitons for which, contrarily to atoms, the Mott transition density is rather close to the typical densities for which BEC is expected to occur. On the other hand, the possibility of achieving a quantum fluid in a solid-state device, with ease of control and integration, would open a new promising way to the implementation of quantum information technology.

Recently, it was suggested that a quantum phase transition of polaritons in a semiconductor microcavity under steady-state incoherent optical pumping might occur, with formation of a collective state of many polaritons. The interest of this system resides in the mixed nature of polaritons, which are a linear superposition of photon and exciton states. Due to the energy-momentum dispersion of a planar microcavity, which is parabolic around zero momentum, the resulting polariton quasiparticle has a very light effective mass at the band bottom, five orders of magnitude smaller than the free electron mass. Another key feature of microcavity polaritons is the very short radiative lifetime, spanning the range of 1 to 10 ps depending on the quality factor of the microcavity. Because of this short lifetime and of the much slower energy-relaxation mechanisms, the polariton system under high energy optical excitation is strongly out of thermodynamic equilibrium. In this situation, the simple picture of an equilibrium Bose gas is completely inadequate. Incidentally, this also implies that the often advocated role of the light polariton mass in determining a high BEC critical temperature, based on the simple equilibrium expression for $T_c$, is actually irrelevant in all experimental situations. The light mass does however play a very important role in three other respects. First, it produces a very long polariton coherence length which averages out the structural disorder of the semiconductor heterostructure thus eliminating the effect of condensate fragmentation. Second, it reduces the final-state phase space available for polariton-polariton scattering processes, resulting in a strong suppression of the dephasing compared to the exciton system, which should play in favor of BEC. Third, for an analogous reason it is at the origin of a very slow condensation kinetics which, in the strongly nonequilibrium condition of polaritons, could play considerably against BEC. This complex scenario suggests that polariton BEC might be possible provided that the density, which is required for the condensation kinetics to be faster than the radiative lifetime, is low enough. It also shows that a reliable theoretical model for polariton BEC must include polariton-polariton scattering and nonequilibrium kinetics on equal grounds.

A parallel between a microcavity polaritons collective quantum state and conventional BEC is made hazardous by the Hohenberg-Mermin-Wagner theorem stating that a phase transition with an evident symmetry breaking is forbidden in a 2-dimensional system. For this reason, the phenomenon has been rather interpreted as a po-
In more practical terms, the BEC scenario is recovered in two dimensions if a finite system size is considered. In this case, condensation occurs because of the finite energy gap, due to energy quantization, between the lowest and the first excited state of the system. This gap quenches the long-wavelength quantum fluctuations which in the limit of infinite size destroy the condensate. For polaritons, a finite quantization size is naturally introduced by polariton localization over a few tens of μm resulting from defects in the microcavity structure or by the finite size of the laser excitation spot. A few experimental results suggest some kind of stimulated phenomenon or even a phase transition with spontaneous phase buildup but many observed features, among which the unexpected observation of a thermal-type two-photon correlation function far above threshold still do not match neither the laser nor the BEC picture.

Laussy et al. have pointed out that an important role is played by the particle number conservation. Indeed, in any symmetry breaking approach, a state with a well-defined quantum phase cannot be stationary, due to the fluctuations of the particle number. Therefore, as in the theory of BEC in diluted atoms, a number-conserving approach is needed in order to correctly describe the quantum phase diffusion of the condensate. To investigate the appearance of condensation (either at finite temperature or in a non-equilibrium regime), it is important to remark that in the BEC models both the condensed and the non-condensed phases, having different fluctuation terms, are considered. In these models, the interactions are the key feature at the origin of the collective excitation spectrum, and are responsible for the scattering kinetics that determines the ratio between condensate and excitation populations. On the theoretical side, many existing works on polaritons prefer overlooking this aspect, pursuing a strict analogy with the laser theory or however neglecting the role of many-body interactions.

In this work we develop a model of the polariton dynamics which includes the polariton-polariton Coulomb interaction and the polariton-phonon scattering on equal grounds, considering a non-equilibrium steady-state optical pump populating the high energy states. The model is derived within the Hartree-Fock-Bogoliubov (HFB) approximation, as in the case of quantum fluids at finite temperature, but imposing the particle number conservation. We introduce the key assumption that the energy-relaxation processes are much slower than the polariton-field dynamics induced by the Coulomb interaction. This adiabatic assumption allows to compute the relaxation kinetics onto a quasistationary HFB spectrum. This situation, which for polaritons is justified by the very slow relaxation kinetics within the steep region of the lower polariton dispersion curve, is exactly opposite to the case of BEC of diluted alkali atoms where the weak mutual interactions ensure a slow field dynamics compared to the thermalization processes. We show that the Coulomb interaction is responsible for a depletion of the polariton condensate even far above threshold, in favour of the excitations. This condensate depletion is strictly related to the existence of collective Bogolubov modes and becomes the dominant process in the limit of large system size, effectively preventing condensation. In Section II we derive the full theoretical formalism and discuss its implications. Section III presents an application to a simplified few-level model, which might describe a situation with polariton lateral confinement and sizeable energy quantization. Section IV is devoted to the discussion of the numerical results. In Section V we present our conclusions and outlook.

II. THEORY

We consider the polariton in the lower branch of the dispersion as a quasi-particle in two dimensions, described by the Bose field ˆp_k:

$$[\hat{p}_k, \hat{p}_{k'}^\dagger] = \delta_{kk'}.$$  \hspace{1cm} (1)

The lower polariton Hamiltonian in presence of Coulomb and polariton-phonon scattering is:

$$H = \sum_k \hbar \omega_k \hat{p}_k^\dagger \hat{p}_k + \sum_q \hbar \omega_q b_q^\dagger b_q + H_C + H_{ph},$$  \hspace{1cm} (2)

$$H_C = \frac{1}{2} \sum_{kk'q} v^{(q)}_{kk'} \hat{p}_{k+q}^\dagger \hat{p}_{k'-q}^\dagger \hat{p}_{k'} \hat{p}_k,$$  \hspace{1cm} (3)

$$H_{ph} = \sum_{kk'q} g^{(q)}_{kk'} (b_q^\dagger + b_{-q}) (\hat{p}_k^\dagger \hat{p}_{k'} + \hat{p}_{k'}^\dagger \hat{p}_k),$$  \hspace{1cm} (4)

where the the matrix element for polariton-polariton interaction $v^{(q)}_{kk'}$ can be derived from the Coulomb interaction between excitons and from the oscillator strength saturation term originating from Pauli exclusion. The polariton-phonon matrix element $g^{(q)}_{kk'}$ can be derived from the deformation potential interaction with acoustic phonons which is expected to dominate at low temperature, but could also include other electron-phonon coupling mechanisms. Since we aim at a kinetic description of the polariton dynamics, we adopt the number-conserving HFB approximation. In fact, although the total number of particles is expected to vary in presence of a pump and of finite escape probability through the mirrors, we still cannot lift the constraint of particle-number conservation, basically for two reasons. First, the description of the condensate as a classical field would result in an unphysical kinetic equation for the condensate, in which the spontaneous in-scattering term vanishes, as shown at the end of this section. Second, at any fixed time the number of particles is well defined in the real system. The energy eigenvalues of the Bogolubov-like excited states depend self-consistently on the actual number of condensed and non-condensed particles.
the energy relaxation rates. One way to overcome the first of the two problems within a symmetry-breaking approach consists in writing a separate semi-classical Boltzmann equation for the condensate density and introducing a stimulation term scaling as the inverse of the area, as was done by Doan et al.\cite{Doan98} Here, however, we prefer to adopt the number-conserving formalism which directly leads to fully self-consistent kinetic equations.

In the number-conserving HFB, the polariton field is expressed as

\[ \hat{p}_k = P_k \hat{a} + \hat{p}_k, \]  

i.e. the sum of a condensate part \( P_k \hat{a} \) and a single-particle excitation part \( \hat{p}_k \). The condensed particle operator obeys the Bose commutation rule \( [\hat{a}, \hat{a}^\dagger] = 1 \) and \( N_c = \langle \hat{a}^\dagger \hat{a} \rangle \) defines the population of condensed particles, while \( P_k \) represents the normalized wave function of the condensate in momentum space. In a non-uniform condensate,\cite{Doan98} this wave function is determined self-consistently by imposing the relation \( \langle \hat{a}^\dagger \hat{p}_k \rangle = 0 \), resulting in the Gross-Pitaevskii equation in the thermodynamic limit. The single-particle excitation \( \hat{p}_k \) is orthogonal to the wave function of the condensate,

\[ \sum_k P_k^* \hat{p}_k = 0, \]

and obeys the modified Bose commutation relation

\[ [\hat{p}_k, \hat{p}_{k'}^\dagger] = \delta_{kk'} - P_k P_{k'}^*, \]

required to preserve the Bose commutation relation for the total field. Using these definitions, the total population of particles with momentum \( k \) is

\[ N_k = \langle \hat{p}_k^\dagger \hat{p}_k \rangle = |P_k|^2 N_c + \hat{N}_k, \]

where \( \hat{N}_k = \langle \hat{p}_k^\dagger \hat{p}_k \rangle \) is the non-condensed population.

The time evolution of the populations can be evaluated by means of the Heisenberg equations of motion. As a first step, we consider only the Coulomb interaction Hamiltonian, neglecting the polariton-phonon scattering. We then obtain the following equations for the dynamics of the field operators:

\[ \dot{\hat{a}} = \sum_k i P_k^* \hat{p}_k + \sum_k P_k^* (\omega_k \hat{p}_k + v \sum_{k', q} \hat{p}_{k' - q}^\dagger \hat{p}_{k'} \hat{p}_{k+q}) \]  

and

\[ \dot{\hat{p}}_k = \omega_k \hat{p}_k + v \sum_{k', q} \hat{p}_{k' - q}^\dagger \hat{p}_{k'} \hat{p}_{k+q} - i P_k \hat{a} - iP_k \hat{a}. \]

Notice that here and in the following we assume a contact polariton-polariton interaction, i.e. \( \psi_{k,k'}^\text{c} = hv \), and we adopt mean-field factorizations, as detailed in the Appendix. Let us introduce the amplitude of the scattering process bringing two particles from the non-condensate to the condensate

\[ \tilde{m}_{k,k'} \equiv \langle \hat{a}^\dagger \hat{a}^\dagger \hat{p}_k \hat{p}_{k'} \rangle. \]

Then the Heisenberg equations result in the following time evolution of the condensate population

\[ \dot{N}_c = -\sum_k \dot{\hat{N}}_k = 2v \text{Im} \left\{ \sum_{k,k',q} P_k'^* \langle \hat{a}^\dagger \hat{p}_{k' - q}^\dagger \hat{p}_k \hat{p}_{k+q} \rangle \right\} \]

\[ = 2v \text{Im} \left\{ \sum_{k,k',q} P_k'^* P_{k' - q} \tilde{m}_{k',k-q} \right\}. \]

In order to simplify the present analysis, we specialize the model to a spatially homogeneous system. In such a limit, the condensate wave function can be safely assumed as a homogeneous function in the spatial domain. We expect the total deviation from this approximation to be small, as the actual wave-function will differ only at the system boundaries. In momentum space, this assumption implies \( P_k = e^{i\phi} \delta_{0,k} \), where \( \phi \) is the condensate macroscopic phase. The assumption therefore implies that the condensate state is always characterized by \( k = 0 \). In the following the macroscopic phase factor \( e^{i\phi} \), together with its time dependence, will be included in the definition of the operator \( \hat{a} \). We point out that the assumption of a homogeneous system is not in contrast with that of finite size, provided the system size is large enough to neglect boundary effects. The state orthogonality in this case implies \( \hat{p}_k = \tilde{p}_k \) for \( k \neq 0 \). As a result, only the diagonal scattering amplitudes \( \tilde{m}_k = \langle \hat{a}^\dagger \hat{a}^\dagger \hat{p}_k \hat{p}_k \rangle \) appear in the equations, in analogy with the anomalous correlations entering the standard HFB approach.\cite{Doan98} In this limit, Eq. \( (12) \) becomes

\[ \dot{N}_c = 2v \text{Im} \left\{ \sum_k \tilde{m}_k \right\} = 2v \text{Im} \{ \tilde{m} \}, \]

where we have defined the total scattering amplitude \( \tilde{m} \) as a sum over all possible final states of the Coulomb scattering process.

Turning to the kinetics of the non-condensate degrees of freedom, we define the destruction operator of a condensate excitation\cite{Doan98}

\[ \hat{\Lambda}_k \equiv \frac{1}{\sqrt{N}} \hat{a}^\dagger \tilde{p}_k, \]

that creates a condensate particle by destroying a non-condensate one. This operator obeys quasi-Bose commutation rules

\[ [\hat{\Lambda}_k, \hat{\Lambda}_q^\dagger] = 0 \]

and

\[ [\hat{\Lambda}_k, \hat{\Lambda}_q] = \delta_{k,q} (N_c - \hat{N}_k - 1)/N. \]

Introducing the standard Bogolubov transformation, the single-particle excitations can be expressed as

\[ \hat{\Lambda}_k = U_k \hat{a}_k + V_k^* \hat{a}^\dagger_{-k}, \]
where $U_k$ and $V_k$ are modal functions, and $\hat{\alpha}_k$ are the operators for Bose normal modes corresponding to the collective excitations of the system. In particular, the commutation rule \footnote{23} impose the condition $U_k V^*_k = U_{-k} V^*_k$ \footnote{27,40}. Using Eq. \footnote{17}, we obtain a direct relation between the one-particle density matrix $(\hat{p}^\dagger_k \hat{p}_k)$ and the populations of Bogolubov modes $\bar{N}_k = (\hat{\alpha}_k^\dagger \hat{\alpha}_k)$:

$$\langle \hat{p}^\dagger_k \hat{p}_k \rangle \sim \delta_{kk'}([|U_k|^2 + |V_k|^2])\bar{N}_k + |V_k|^2].$$ \hspace{1cm} (18)

This brings to the result, expected by symmetry arguments, that for a spatially homogeneous system the off-diagonal density matrix terms of the non-condensed states are vanishing within the mean-field approach, i.e.

$$\bar{N}_{k,k'} \equiv \langle \hat{p}^\dagger_k \hat{p}_{k'} \rangle = \bar{N}_k \delta_{kk'}.$$ \hspace{1cm} (19)

Hence, we obtain the following equation for the population of the excited states

$$\dot{\bar{N}}_k = -2v\text{Im}\{\hat{m}_k - \sum_{q} (\hat{p}^\dagger_q \hat{p}_q \bar{N}_k - \hat{p}_q \hat{p}^\dagger_q \bar{N}_k - k \cdots k')\}. \hspace{1cm} (20)$$

Finally the scattering amplitude $\hat{m}_k$ obeys the equation (see the Appendix for the derivation)

$$\dot{\hat{m}}_k = \iota \Omega_k \hat{m}_k - 2iv\bar{N}_k \hat{m}_k - iv(1 + 2\bar{N}_k) N_c (N_c - 1) + iv(1 + 2N_c) \sum_{q} (\hat{p}^\dagger_q \hat{p}_q \bar{N}_k \hat{p}_k).$$ \hspace{1cm} (21)

where $\Omega_k = -2[\omega_k + v(N_c - \bar{N}_k - 5/2)].$

The residual two-particle correlations for excited particles, appearing in Eqs. \footnote{20} and \footnote{21}, require special care as they are the source of the off-diagonal long-range correlations characterizing a Bose-Einstein condensate in real space. Once again, these terms in the number-conserving approach are analogous to the anomalous correlations appearing in a standard HFB formalism. In particular, they cannot be factored in the single-particle basis without affecting the spatial correlation properties of the condensate. We will see in the next section how these terms can be treated within a simplified few-level model. Notice that the quantity $\hat{m}_k$ denotes a scattering process which would not conserve energy in a single-particle picture, as it describes the scattering of two particles from one state to another one at larger energy. This process is actually present in our formalism because the spectrum of the system is modified by the interactions and the new eigenstates are the collective Bogolubov excitations, describing condensate fluctuations with large wavelength, rather than the single-particle states.

We now introduce the contribution to the population kinetics due to polariton-phonon scattering. In the limit of a spatially homogeneous system, we can write the phonon contribution to the population kinetics of the condensate as

$$\dot{N}_c|_{ph} = -\frac{i}{\hbar} \sum_{q,k} g_{c(k)}^q \langle (b_q^\dagger + b_{-q})(\hat{\alpha}^\dagger \hat{p}_k - \hat{p}_k^\dagger \hat{\alpha}) \rangle.$$ \hspace{1cm} (22)

The Heisenberg equations for the operators produce again a hierarchy of equations for phonon-assisted correlations of all orders, which are coupled to the HFB variables. The equations for the first order phonon-assisted correlations, like $g_{c,k} = \langle b_k^\dagger a^\dagger \rangle$, are formally solved within the self-consistent Markov approximation. In particular, the higher order phonon assisted correlations entering the kinetic equations are factored according to the mean field approximation. For example

$$\langle \hat{a}^\dagger \hat{a} \hat{b}^\dagger \hat{p} \rangle \sim (N_c - 1)b_k^\dagger \hat{a}^\dagger \hat{p}_k$$ \hspace{1cm} (23)

or

$$\langle b_{-q} b_k^\dagger \hat{a}^\dagger \hat{p} \rangle \sim \delta_{q,q'k} \delta_{kk'} \bar{N}_k (1 + n_q),$$ \hspace{1cm} (24)

where $n_q$ is the phonon distribution at wave vector $q$. In this way, we obtain equations of the following kind

$$i\partial_t \langle b_q^\dagger \hat{a}^\dagger \hat{p} \rangle = \left[\omega_q + v(N_c + \bar{N}_k - 1) - w_q \right] \langle b_q^\dagger \hat{a}^\dagger \hat{p} \rangle + \frac{g_{c(q)}^q}{\hbar} \left[ (N_c - \bar{N}_k) N_c - (1 + n_q) \bar{N}_k \right],$$ \hspace{1cm} (25)

whose formal solution can be plugged into the HFB equations. The resulting contribution to the dynamics of the condensate population is:

$$\dot{N}_c|_{ph} = 2\pi \sum_{q,k} \delta \left[ \omega_q + v(N_c + \bar{N}_k - 1) - w_q \right] \times \frac{|g_{c(q)}^q|^2}{\hbar} \left[ (\bar{N}_k - n_q) N_c + (1 + n_q) \bar{N}_k \right]$$ \hspace{1cm} (26)

$$= 2 \sum_{k} g_c^{(k)} \left[ (\bar{N}_k - n_c^{(k)}) N_c + (1 + n_c^{(k)}) \bar{N}_k \right],$$ \hspace{1cm} (27)

where $g_c^{(k)}$ and $n_c^{(k)}$ are respectively the effective phonon scattering matrix element and the phonon population at the wave vector defined by momentum and energy conservation. In particular, the in-plane wave vector component $q_{\parallel}$ is fixed by momentum conservation, while the $z$-component $q_z$ is selected by energy conservation. In this way, the polariton-phonon coupling introduces effective phonon-mediated polariton-polariton interaction terms, the lowest-order ones being proportional to $|g_{k'}^{(q)}|^2$. The phonon populations $n_q$ are assumed to be thermally distributed at the lattice temperature. Eq. \footnote{20} is the standard Boltzmann equation expected for the energy-relaxation kinetics. A similar Boltzmann equation holds for the populations $\bar{N}_k$ of the excited single-particle states. The phonon-assisted correlations entering the equations for the scattering amplitudes $\hat{m}_k$, on the other hand, cannot be solved analytically as was done for Eq. \footnote{26}, because Coulomb interaction couples different values of the momentum.

We conclude this section by giving the explicit expression of the phonon coupling term, as it would appear in the equation for the condensate if a symmetry-breaking approach was adopted. Expressing the polariton field as

$$\hat{p}_k = P_k + \hat{p}_k,$$ \hspace{1cm} (27)
where $P_k = \langle \hat{p}_k \rangle$ is a classical field, while $\hat{p}_k$ describes fluctuations, and considering again the uniform limit $P_k = \delta_{k,0} P$, the phonon contribution to the condensate field equation reads

$$\hat{P} \big|_{ph} = -\frac{i}{\hbar} \sum_{q,k} g_{k0}^{(q)} ((\hat{b}_q^{\dagger} + b_{-q}) \hat{p}_k). \tag{28}$$

Following the same procedure adopted above in order to evaluate the phonon assisted terms within the Markov approximation, we obtain for the condensate population $N_c \equiv |P|^2$ the following expression

$$\hat{N}_c \big|_{ph} = 2\pi \sum_{q,k} \delta(\omega_k - \omega_q) \left| g_{k0}^{(q)} \right|^2 (\hat{N}_k - n_q) N_c, \tag{29}$$

Where the in-scattering term present in Eq. (29) is here instead missing. This term, responsible for the spontaneous scattering into the condensate, is of course absent within a description of the condensate as a classical field. As suggested at the beginning of this section, the adoption of the number-conserving formalism prevents the occurrence of this unphysical behavior.

III. FEW-LEVEL MODEL

As seen in the previous Section, the solution of the whole set of equations for the populations and correlations is a challenging task, basically because of the off-diagonal coupling in the phonon assisted correlations. In addition, the two-point correlations between single-particle excitations appearing in Eqs. (20) and (21) still need to be addressed in a consistent way. In a typical photoluminescence experiment under nonresonant excitation, the steep polariton dispersion results in a relaxation bottleneck\[24,51\] with polariton population piling up at the boundary of the flat exciton-like region of the polariton dispersion. From there, polaritons relax to the band-bottom, where the actual phase transition can take place before they recombine emitting a photon. Given the very slow relaxation rate\[24\] and the very fast radiative recombination rates, the relaxation from the bottleneck region to the polariton band bottom is very likely to take place in one, or at most a few relaxation steps. Multiple relaxation steps within the steep region of the polariton dispersion are however very unlikely. Their contribution might quantitatively affect the total out-scattering rate from the bottleneck region, but it will not affect in a sizeable way the in-scattering rate in the ground level, which is the relevant process for final-state stimulation. This allows the introduction of an effective-level scheme, which should describe the relevant dynamics of the problem. In this scheme, the flat bottleneck region of the dispersion, the excited states within the steep region and the ground state are described as three effective levels, accounting for the respective density of states in the real system. We will refer to these levels as bottleneck, single-particle excitations and condensate respectively. For the reasons illustrated above, we can assume that the phonon mediated relaxation occurs only between the bottleneck level and the two other levels, while the Coulomb amplitudes $\tilde{n}_k$ are non-vanishing only for states energetically close to the condensate, i.e. in the band bottom region. The first assumption, as already stated, stems from the fast radiative rate of the polariton levels in the strong coupling region of the dispersion, ensuring a negligible contribution to the in-scattering rate in the ground level\[24,25\]. The second assumption is justified by the fact that the Coulomb scattering amplitudes $\tilde{n}_k$ can be important only for small wave vector $k$, because they are linked to condensate fluctuation of large wavelength, as discussed previously. This scheme is analogous to the one commonly adopted for the description of BEC kinetics of a diluted alkali gas\[24,25\]. The idea behind it is that in the lower energy region of the spectrum, the Bogolubov field dynamics is the dominant process, while the low density of states makes the relaxation kinetics negligible. The opposite occurs in the higher-energy region, where most of the relaxation kinetics takes place but the collective Bogolubov excitations coincide with the single-particle states\[24,25\]. Our simplified few-level model is sketched in Fig. 1. The creation operator for the bottleneck state is defined by $\hat{p}_k^{\dagger}$ (and $N_1$ is the population per mode at bottleneck), while $\hat{P}$ now indicates the creator for a single-particle excitation. The corresponding total population of single-particle excitations is given by $\hat{N} = \sum_k \hat{N}_k$. Our purpose is to write a set of equations giving the time evolution of the condensate population $N_c$, of the single-particle excitations population $\hat{N}$ and of the bottleneck population $N_1$.

As in previous treatments\[53\], the introduction of effective levels requires the introduction of renormalized coupling constants. In our case, the effective phonon coupling rates involving the bottleneck level are proportional to the total number of states $\rho_x$ in this region of the dispersion. This latter is estimated from the assumption of a thermalized polariton distribution at the bottleneck\[24\] and is related to the exciton mass $M_{exc}$ and the exciton energy thermal broadening $E \approx k_B T$ by

$$\rho_x = \left(\frac{A}{2\pi}\right) (M_{exc} E/\hbar^2), \tag{30}$$

resulting in an effective phonon coupling rate $g_k = \rho_x g_k^{(1)}$, where $g_k^{(1)}$ is the phonon coupling rate for a scattering process between a state at the bottleneck and a polariton state with momentum $k$, defined as in equation (20)\[24,25\].

We now address the problem of the two-particle correlation terms entering equations (20) and (21). Using again Bogolubov transformations\[17\], linking the single-particle field to collective excitations, we can write

$$\sum_{q,k} \langle \hat{p}_q \hat{p}_q^{\dagger} \hat{p}_k \hat{p}_k \rangle = \Upsilon(N) \left[ \sum_k U_k V_k^* (1 + 2 \tilde{N}_k) \right]^2$$
The amplitude of this term depends on the anomalous coefficients $V_k$, which tend to zero for vanishing condensate density. Therefore, in the thermodynamic limit, since the quantities $\bar{N}_c$ and $V_k$ vanishes as expected for a Bose system in two dimensions, the quantity in Eq. (31) reduces to a term proportional to $\bar{n}\bar{N}$, where $\bar{n}$ is the particle density. On the other hand, in order to solve the effective-level equations presented below, we want rewrite this contribution in a more useful k-independent form. To this purpose, we propose the following argument based on the two-point spatial correlation function. In real space, we can write

$$\sum_{q, k} \langle \hat{p}_q^\dagger \hat{p}_q \rangle \sim \int d\mathbf{r} ds \langle \hat{p}_1^\dagger (\mathbf{r}) \hat{p}_1 (\mathbf{r}) \hat{p}(s) \hat{p}(s) \rangle. \quad (35)$$

Now, for a non-condensed system, at the lowest order in the total density, the four-point spatial correlation can be safely factorized in terms of the two-point correlation $\langle \hat{p}_1^\dagger (\mathbf{r}) \hat{p}_1 (\mathbf{r}) \rangle$, as

$$\langle \hat{p}_1^\dagger (\mathbf{r}) \hat{p}_1 (\mathbf{r}) \hat{p}(s) \hat{p}(s) \rangle \sim 2 \langle \hat{p}_1^\dagger (\mathbf{r}) \hat{p}_1 (\mathbf{r}) \rangle^2 - \delta(\mathbf{r} - \mathbf{s}) \bar{N}(\mathbf{r}). \quad (36)$$

Therefore, defining the parameter

$$\alpha \equiv \bar{N}^{-2} \int d\mathbf{r} ds \langle \hat{p}_1^\dagger (\mathbf{r}) \hat{p}(s) \rangle^2, \quad (37)$$

we obtain

$$\sum_{q, k} \langle \hat{p}_q^\dagger \hat{p}_q \rangle \sim (2\alpha \bar{N} - 1) \bar{N}^{-1}. \quad (38)$$

In an equilibrium situation, for a non-condensed phase, the two-point spatial correlation vanishes at distances larger than the thermal length $\lambda_T = \sqrt{2\hbar^2/(mk_BT)}$, which derives directly from the equilibrium Bose-Einstein distribution of excitations. In a non-equilibrium situation like the present one, the distribution of excitations generally differs from the equilibrium one, but again it corresponds to a characteristic length. We however expect the correlation length $\kappa$ of the one-body density function, defining the spatial correlation length for a non-condensed system, to be in the same range as $\lambda_T$. Therefore, from Eq. (37) we see that

$$\alpha \sim \frac{\kappa^2}{\lambda_T^2}. \quad (39)$$

Assuming $\kappa$ independent of the system size, $\alpha$ then scales as $A^{-1}$. For a system that has undergone condensation, on the other hand, the simple factorization used in Eq. (36) is not valid, as it totally neglects the anomalous correlations. These contributions are the dominant term in Eq. (31), proportional to the products $U_k V_k^*$. Inspection of Eq. (31), shows that its dominant term still depends on the squared population of excitations $\bar{N}^2$, as previously discussed. We therefore propose to use the expression (38) also in the condensate regime, by making a different assumption on the parameter $\alpha$. In particular, for a condensed system, the parameter $\alpha$ should no longer scale as in Eq. (39), because of the presence of long-range correlations. At the same time, comparing equations (38) and (39), we see that only values $\alpha < 1$ are admitted, the limiting value $\alpha = 1$ corresponding to a four-point spatial correlation function extending over the whole system size. We still expect, however, a residual dependence of $\alpha$ on the system area $A$ even in the...
condensed regime, because by increasing the system size we suppress the relaxation mechanism in favour of the condensate depletion. Therefore, a self-consistent evaluation of the parameter $\alpha$ is needed for the present kinetic model to have the correct thermodynamic limit. We discuss this approach below, after having introduced the effective-level description of the system. Concluding this analysis, we remark that the quantity $N_c$ is real, so the last term in equation (20) gives a vanishing contribution when summed over $k$.

In order to obtain a closed set of equations for $N_c$, $\tilde{N}$, $N_1$ and $\tilde{m}$, the energies of the single-particle excitations are replaced by an effective value, $\hbar \tilde{\omega} \approx \hbar \omega$, representing a typical energy of the non-condensed levels relative to the ground state. This quantity plays a crucial role in this effective-level scheme, as it determines the finite energy gap that makes condensation possible in a two-dimensional system. We discuss below how the numerical value of this parameter is chosen. Similarly, we introduce the energy $\hbar \omega_1$ for the bottleneck states. We assume intrinsic linewidths $\hbar \gamma_1$ for the bottleneck level and $\hbar \gamma_0$ for the other levels, accounting for radiative recombination as well as nonradiative homogeneous energy broadening. In this way, using the relation (38), the resulting equations are

$$\dot{N}_c = -2\gamma_0 N_c + 2g_c \Gamma (1 + n_c) N_1 (1 + N_c) - 2g_c \Gamma n_c (1 + N_1) N_c + 2\text{Im}\{\tilde{m}\},$$

$$\dot{\tilde{N}} = -2\gamma_0 \tilde{N} + 2g_1 \Gamma (1 + \tilde{n}) N_1 (\eta + \tilde{N}) - 2g_1 \Gamma \tilde{n} (1 + N_1) \tilde{N} - 2\text{Im}\{\tilde{m}\},$$

$$\dot{N}_1 = -2\gamma_1 N_1 + 2\Gamma (g_c n_c N_c + g_1 \tilde{n} \tilde{N}) (1 + N_1) - 2\Gamma \left[ g_c (1 + n_c) (1 + N_1) + g_1 (1 + \tilde{n}) (\eta + \tilde{N}) \right] N_1 + F,$$

$$\dot{\tilde{m}} = -2 \left\{ 2\gamma_0 + \Gamma_m \left[ g' (n' - N_1) + g'' (n'' - N_1) \right] + \text{Im} \{ Y \} \right\} \tilde{m} + iv \left[ (1 + 2N_c) \tilde{N} (2\alpha \tilde{N} - 1) - (\eta + 2\tilde{N}) N_c (N_c - 1) \right],$$

where $\Gamma = \gamma_1 / (\gamma_1 + \gamma_0)$, $\Gamma_m = \gamma_1 / (\gamma_1 + 3\gamma_0)$, $\Omega' = \sqrt[2] {\omega + v (N_c - (1 - 1/\eta) N - 5/2)}$ and $\eta$ is the number of single-particle excitation states, depending on the quantization area via the relation $k = (n_x x + n_y y) \pi / \sqrt{\Omega'$. We have denoted by $n_c$, $\tilde{n}$, $n'$ and $n''$ the phonon populations defined by energy conservation within the Markov assumption, as shown in equation (20). Correspondingly, $g_c$, $g_1$, $g'$ and $g''$ denote the effective phonon coupling strength at the same energies, renormalized by the number of bottleneck levels $\rho_x$. The equation for the scattering amplitude contains an oscillating term, whose frequency depends on the actual energy needed to create condensate particle. Notice that all this quantities depend on the actual condensate and non-condensate densities and vary self-consistently during time evolution. In particular, the parameter $\alpha$ can be obtained self-consistently at each step of the kinetics, by equating expressions (34) and (38). The effective-level representation of the condensate excitation naturally implies an effective Bogolubov factor $V$, via Eq. (38), and corresponding values for the other related quantities. The expression for $\alpha$ takes then the compact form

$$\alpha(N) = \left. \frac{1 + N_c}{2 + N_c} \left( \frac{1}{\eta} + 2 \frac{[U^* V]^2}{\lambda^2} \right) \right), \quad (41)$$

In particular, we find again that the first term in parenthesis scales as $A^{-1}$, consistently with the previous discussion. Actually, it turns out that the precise value of $\alpha$ is not critical in determining the condensation dynamics, provided that the two limits $\alpha = 0$ and $\alpha = 1$ are not reached. In particular, in the next section, we will show how the self-consistent result does not differ much by one one obtained using a constant value for $\alpha$. We have introduced a steady-state pump rate $F$ in the equation for the effective bottleneck level. This quantity represents the number of particles per unit time and per state, which enters the system following the relaxation from higher-energy states, as in a typical experiment with non-resonant continuous-wave excitation. For a given total pump flux $j$ (total rate of polaritons per unit area), the quantity $F$ can be rewritten as $F = j A \rho_x^{-1}$. This shows that for a given pump flux the quantity $F$ does not depend on the area $A$ of the system, as can be argued from Eq. (30).

The phonon-mediated interaction results in a Boltzmann dynamics with scattering coefficients $g_c \Gamma$, $g_1 \Gamma_m$, $g' \Gamma_m$ and $g'' \Gamma_m$. In the kinetics described by our model, the initial growth of the condensate is triggered by final-state stimulation within the relaxation process. It takes place in the ground state which is separated by an energy gap $\tilde{\omega}$ from the excited level. Above the stimulation threshold, a crucial role is played by the Coulomb scattering amplitude $\tilde{m}$, which enters the first two equations in (41) with equal factors up to a sign, and determines the actual condensate growth or depletion. As we will see, this term determines the macroscopic condensate fraction reached by the system. Indeed, we can
compute the approximate analytical steady state fraction, 

depletion mechanism. sets a lower bound to the effectiveness of the condensate with respect to our simplified assumption which therefore ply an increase of the total coherent outscattering rate. We area and making the high density limit \( \tilde{\alpha} \)\,\,\,\,(2)\,\,\,\,\,

equation for \( \gamma_1 \)\,\,\,\,\,

variation of the relaxation rates) and in the occurrence results both in a modified spectrum (with subsequent density at bottleneck and to the condensate density, as would not introduce significant changes to the exciton density. Prediction the coexistence of condensate and non-condensate even far above threshold. This behaviour, as will be also seen in the numerical solution, originates exclusively from the Coulomb scattering, whereas a pure Boltzmann dynamics would always predict a condensate fraction approaching 1 above threshold.

The adoption of this simplified model calls for some additional remarks, in order to understand its limitations and the relevance of the parameters. As already argued, we can expect that the relaxation from the bottleneck region into the low-energy states is qualitative well described within a few-level approximation, as the inclusion of other intermediate states would only result in a finite increase of the pump threshold. This in turn would not introduce significant changes to the exciton density at bottleneck and to the condensate density, as they mainly depend on the phonon coupling rates for the direct scattering processes. The Coulomb interaction results both in a modified spectrum (with subsequent variation of the relaxation rates) and in the occurrence of coherent scattering processes between the condensate and the states close to zero momentum. Both effects are mainly important for the lowest-lying states. In order to correctly describe the dominant role played by the low-energy states, the parameter \( \hbar \omega \) has to be set to a value lower than a simple average of the excited energy eigen-values. This parameter represents in our model the energy gap between condensate and excited states. It thus plays the same role as the finite-size energy quantization in a fully two-dimensional system of finite size. We follow this prescription and therefore set the parameter \( \hbar \omega \) to the energy gap that would result from the system area \( A \). The presence of more excited levels would imply an increase of the total coherent outscattering rate with respect to our simplified assumption which therefore sets a lower bound to the effectiveness of the condensate depletion mechanism.

IV. NUMERICAL SOLUTION

For the numerical evaluation, parameters of a typical AlGaAs microcavity with one embedded GaAs quantum well have been used, with a quantization area \( A = 100 \mu m^2 \). The quantity \( A \) enters the definition of the phonon coupling term as well as the expression for \( \tilde{\rho} \) and implies a number of excited levels \( \eta = 30 \) for which we take the representative energy \( \hbar \omega = 0.1 \) meV (see the discussion in the previous section). For these parameters, the effective phonon coupling strengths linking the bottleneck and the low-lying states are \( \gamma_c, \tilde{\gamma} \approx 1 \mu eV \), resulting in a very long relaxation time \( (\tau_{relax} \approx 100 \) ps), consistently with the adiabatic assumption that we have made. The Coulomb matrix element is evaluated to be \( \hbar \omega = 5 \times 10^{-4} \) meV. The other parameters are \( T = 10 \) K, \( \hbar \gamma_0 = 0.2 \) meV, \( \hbar \gamma_1 = 1 \) meV, \( \hbar \Omega_R = 3.5 \) meV, \( \hbar (\omega_1 - \tilde{\omega}) = 1.9 \) meV and \( \rho_x = 10^4 \). The value \( \hbar \gamma_0 = 0.2 \) meV is typical for the radiative linewidth of a polariton at zero exciton-cavity detuning in such a system. The value \( \hbar \gamma_1 = 1 \) meV for the exciton-like part of the polariton branch, on the other hand, accounts for the density-dependent exciton dephasing rate expected in the vicinity of the transition density resulting from our calculations. We have solved numerically the set of equations as a function of time. For each value of the pump density \( f \), we observe a time-dependent transient followed by a stationary solution for all the quantities. Fig. 2(a) displays the stationary populations as a function of \( f \). In order to check that the choice of parameter \( \alpha \) is not critical, we compare the results obtained using a constant value \( \alpha = 0.4 \) with the results obtained calculating self-consistently \( \alpha \) from Eq. (11), at each step of the relaxation. The figure clearly shows that the two methods give very similar results. Obviously, by using a density-independent \( \alpha \), the condensate fraction at low pump density is overestimated). Below threshold, Boltzmann relaxation results in an increase of the polariton population, in which the condensate population remains microscopic, i.e. \( N_c \ll N \). A threshold occurs at \( f = f_{th} \), for which \( N_c \simeq 1 \). Above threshold, the bottleneck population \( N_1 \) reaches a saturation value while the condensate population \( N_c \) becomes a macroscopic fraction of the non condensed polariton population \( N \), which in turn continues to grow with \( f \). The behaviour of the ratio \( N_c/N \) is plotted in Fig. 2(b). At \( f \gg f_{th} \) the condensate fraction approaches a finite value lower than 1, consistent with \( \alpha = 0.4 \). Fig. 2(c) displays the imaginary part of the steady-state Coulomb scattering amplitude \( \tilde{\alpha} \). Close to threshold, in correspondence to low values of the condensate fraction, this quantity takes positive values, thus favoring condensation. Above threshold, on the other hand, it takes large negative values, resulting in condensate depletion. Hence, Coulomb interaction plays a crucial role during phase transition, as expected according to both laser and BEC quantum theories. In order to clarify the effect of the Coulomb interaction, we compare in Fig. 2(a) and (b) the steady state solutions obtained neglecting all Coulomb terms in 10. By inspection of Eqs. 10, it is clear that in this case the populations obey a standard Boltzmann dynamics. Without Coulomb interaction, therefore, a standard
three-level Boltzmann equation is recovered. At threshold, the condensate population starts to grow due to final state stimulation and the system undergoes a complete transition to a fully condensed regime with a condensate fraction equal to 1.

The threshold $f_{th}$ depends on the energy gap $\bar{\omega}$. In the limit of a system of infinite size, this energy gap vanishes and $f_{th}$ becomes infinite. In order to understand how the size of the system affects the condensate fraction, on the other hand, in Fig. 3 we compare the results obtained with different values of $A$. For this comparison, we assume that $\bar{\omega}$ scales as $A^{-1}$, according to the energy quantization in a quantum system of finite size. The results plotted in Fig. 3 show that the condensate fraction reached far above threshold depends on the system size. This condensate depletion is due to the coherent scattering terms out of the condensate that originate from the kinetic HFB equations. In the thermodynamic limit, therefore, our model predicts a vanishing condensate fraction, as imposed by the Hohenberg-Mermin-Wagner theorem. If in the present model we neglect the Coulomb interaction, the coherent scattering terms are absent, resulting in a condensate fraction that always tends to 1 far above the threshold pump intensity (see Fig. 3 (b)). The present kinetic model therefore correctly reproduces the behaviour of a two-dimensional Bose system in the thermodynamic limit, where the long-wavelength fluctuations are expected to destroy the condensate.

To better understand the interplay between scattering amplitudes and condensate growth, we display the time evolution of $N_c$ and $N$ in Fig. 3(a) and of the imaginary part of $\tilde{m}$ in Fig. 3(b). At short times, the quantity have not a stationary value, due to the relaxation dynamics and the condensate population is small. Correspondingly, the quantity $\text{Im}\{\tilde{m}\}$ takes positive values. At longer times, when all the quantities reach a stationary value, with a macroscopic occupation of the condensate, $\text{Im}\{\tilde{m}\}$ has turned to negative values, implying the resulting condensate depletion.

The Coulomb interaction is therefore the key mechanism determining the condensate fraction under steady-state conditions. For the parameters used in Fig. 3 the saturation value of $N_1$ corresponds here to a bottleneck polariton density $n_B \sim 2 \times 10^{11}$ cm$^{-2}$, larger than the optical saturation density, suggesting that condensation cannot occur in a simple system with one quantum well. When using the parameters of the experiment by Deng et al., i.e. considering a sample containing 12 quantum wells at $T = 4K$ with $\hbar \Omega_R = 7.5$ meV, we obtain a qualitatively similar behaviour (not shown) with $N_1 = 20$ at threshold, resulting in a bottleneck polariton density per quantum well $n_B \sim 7 \times 10^9$ cm$^{-2}$, lower than saturation density and in fairly good agreement with the experimen-
We argue that, in the experiment by Deng et al., the unusually high values measured for the two-photon correlation function might be explained by the coexistence of condensed and non-condensed phases even far above threshold.

V. CONCLUSIONS

We have presented a kinetic theory of microcavity polariton condensation. The theory is based on a number-conserving HFB description of the polariton quantum field, treated using a density-matrix formalism. This allows inclusion of various scattering processes, inducing the energy relaxation and the final-state bosonic stimulation which causes the condensate to grow. In particular, we describe the polariton-phonon scattering, but other scattering mechanisms could in principle be included. Differently from BEC kinetic models in atomic physics, here the HFB field dynamics is much faster than the relaxation kinetics. As a consequence, the steps of the relaxation kinetics follow adiabatically the spectrum of collective modes stemming from the HFB field equations. We show that in this non-equilibrium regime, the condensate fluctuations strongly influence the phase transition even far above threshold. In particular, they induce scattering from the condensate to the excitation modes that can result in a small condensate fraction in presence of slow energy relaxation rates.

Within a few-level model, we have performed a numerical evaluation of the condensation kinetics. It turns out that in realistic experimental conditions the condensate fraction above threshold approaches asymptotically a value significantly lower than 1, depending on the system size, as expected for a two-dimensional Bose system. The condensate fraction can even become vanishing if the size exceeds a few tens of μm.

We conclude that the coexistence of condensate and non-condensate, caused by the Coulomb interaction, is a dominant aspect of the polariton condensation dynamics. This feature, unpredicted by models based on a standard Boltzmann kinetic approach, can affect strongly the coherence properties of condensed polaritons and possibly prevent condensation in the most common experimental conditions. This result holds great importance in the light of the numerous experimental claims of polariton Bose-Einstein condensation and of the recent achievements in lateral confinement of microcavity polaritons over the micrometric scale.

VI. APPENDIX

The equations of motion appearing in this paper are derived in the mean-field limit, i.e. factoring higher order correlation terms in single-particle or Bogolubov quasiparticle population terms. In this appendix we give the details of this prescription. Let us consider the system having a defined number of condensed and non-condensed particles, $N_c$ and $\tilde{N} = \sum \tilde{N}_k$ respectively. We compute the following expectation values of the two-particle quantities

$$\langle \hat{a}^\dagger \hat{a}^\dagger \hat{a} \hat{a} \rangle \simeq N_c (N_c - 1)$$

and

$$\sum_{k\neq q} \langle \hat{p}^\dagger_k \hat{p}^\dagger_{k'} \hat{p}_q \hat{p}_{q'} \rangle \simeq 2\tilde{N}^2 - \tilde{N} - \sum_k \tilde{N}_k^2.$$ (45)

Consistently, we can introduce an approximated Hamiltonian in terms of linearized operators, having the same expectation values as obtained in mean-field limit. The linearized operators are

$$\hat{a}^\dagger \hat{a} \simeq (N_c - 1) \hat{a}^\dagger,$$

$$\hat{a}^\dagger \hat{a} \hat{a} \simeq N_c \hat{a},$$

$$\hat{p}^\dagger_{kq} \hat{p}^\dagger_{k'q'} \hat{p}_q \hat{p}_{q'} \simeq \tilde{N}_{k,q} \hat{p}^\dagger_{k'q'} \hat{p}_q \hat{p}_{q'} + \tilde{N}_{k,q} \hat{p}^\dagger_{k'q'} \hat{p}_q \hat{p}_{q'} - \delta_{k,k'} \delta_{q,q'} (N_k + 1) \hat{p}^\dagger_k \hat{p}_k,$$

$$\hat{p}^\dagger_{kq} \hat{p}^\dagger_{-k'q'} \hat{p}_q \hat{p}_{q'} \simeq (\tilde{N}_{q,k} - \delta_{q,k}) \hat{p}^\dagger_{-q} \hat{p}_q + \tilde{N}_{q,k} \hat{p}^\dagger_{q'} \hat{p}_{q'},$$

$$\hat{p}^\dagger_{q+q'} \hat{p}^\dagger_{-kq} \hat{p}_q \hat{p}_{q'} \simeq \tilde{N}_{q+q'-k,q} \hat{p}^\dagger_{q} \hat{p}_q + \tilde{N}_{q+q'-k,q} \hat{p}^\dagger_{q'} \hat{p}_{q'},$$

$$\hat{p}^\dagger_{kq} \hat{p}^\dagger_{-q} \hat{p}_q \hat{p}_{q'} \simeq \tilde{N}_{k,q} \hat{p}^\dagger_{-q} \hat{p}_q + \tilde{N}_{k,q} \hat{p}^\dagger_{q'} \hat{p}_{q'} - \delta_{q,q'} \tilde{N}_k \hat{p}_k.$$ (51)

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These relations can be used to factor the correlations entering the equation for the scattering amplitudes $\tilde{m}_k$

$$\tilde{m}_k = -2i(\omega_k - \omega_0 - 2v)\tilde{m}_k + v\langle \hat{a}^\dagger \hat{a}^\dagger \hat{a} \hat{a}^\dagger \hat{p}_k \hat{p}_k \rangle_k$$

$$- 4v \sum_q \langle \hat{a}^\dagger \hat{a} \hat{p}_q^\dagger \hat{p}_q \hat{p}_k \hat{p}_k \rangle - v \sum_q \langle (\hat{a}^\dagger \hat{a}) \hat{p}_q^\dagger \hat{p}_q \hat{p}_k \hat{p}_k \rangle + v \langle \hat{a}^\dagger \hat{a} \hat{a} \hat{a}^\dagger \hat{p}_q^\dagger \hat{p}_q^\dagger \hat{p}_k \hat{p}_k \rangle.$$ (52)

In detail, we apply the following factorizations

$$\langle \hat{a}^\dagger \hat{a} \hat{a}^\dagger \hat{a} \hat{p}_k \hat{p}_k \rangle \simeq (N_c - 2)\tilde{m}_k$$ (53)

$$\langle \hat{a}^\dagger \hat{a} \hat{a} \hat{a}^\dagger \hat{p}_k \hat{p}_k \rangle \simeq (N_c - 1)\tilde{m}_k$$ (54)

$$\langle \hat{a}^\dagger \hat{a} \hat{p}_q^\dagger \hat{p}_q \hat{p}_k \hat{p}_k \rangle \simeq (N_q - \delta_{q,k} - \tilde{N}_{-k} - \tilde{N}_{-k})\tilde{m}_k + N_q \tilde{m}_q, -k + \tilde{N}_{-k} \tilde{m}_{-q,k}$$ (55)

$$\langle (\hat{a}^\dagger \hat{a} \hat{a}) \hat{p}_q^\dagger \hat{p}_q \hat{p}_k \hat{p}_k \rangle \simeq (1 + 2N_c)\langle \hat{p}_q^\dagger \hat{p}_q \hat{p}_k \hat{p}_k \rangle.$$ (56)

$$\langle \hat{a}^\dagger \hat{a} \hat{a} \hat{a}^\dagger \hat{p}_k \hat{p}_k \hat{p}_k \hat{p}_k \rangle \simeq N_c (N_c - 1) (1 + \tilde{N}_{-k} + \tilde{N}_{k}).$$ (57)

$$\langle \hat{a}^\dagger \hat{a} \hat{p}_q^\dagger \hat{p}_q \hat{p}_k \hat{p}_k \rangle \simeq 2N_q + q', -k \tilde{m}_q, -k + \tilde{N}_{q', -k} \tilde{m}_{q', -k} + \tilde{N}_{q', -k} \tilde{m}_{q, -k}$$ (58)

$$\langle \hat{a}^\dagger \hat{a} \hat{p}_q^\dagger \hat{p}_q \hat{p}_k \hat{p}_k \rangle \simeq 2N_q + q', -k \tilde{m}_q, -k + \tilde{N}_{q', -k} \tilde{m}_{q', -k} - \delta_{q', -k} \tilde{m}_{q, -k} \tilde{m}_{q', -k}.$$ (59)

Notice that we can also rewrite the double sums as

$$\sum_{qq'} N_{q', -k} \tilde{m}_{q', -k} = \sum_{q', k} N_{q', k} \tilde{m}_q, -k + \sum_{q', k} N_{q', k} \tilde{m}_k$$

$$- N_{k} \tilde{m}_k + \sum_{q', k} N_{q', k} \tilde{m}_{q', -k}.$$ (60)

and the last term can be neglected, within the assumption of a spatially homogeneous system. In this way, Eq. 21 is recovered.
This condition is one of the two entering the standard Bogolubov theory as well. The second one is $|u_k|^2 - |v_k|^2 = 1$ and results from the commutation relation $[\hat{\Lambda}_k, \hat{\Lambda}_q^\dagger]$. In the present treatment, $[\hat{\Lambda}_k, \hat{\Lambda}_q^\dagger] = ([\delta_{k,q} - P_k^* P_q](\hat{a}^\dagger \hat{a} - 1) - \hat{p}_k^* \hat{p}_q)/N$. The result of the standard Bogolubov theory is then recovered by taking the limits $\hat{a}^\dagger \hat{a} \simeq N_c$ and $\hat{p}_k^* \hat{p}_q \simeq \tilde{N}_{q,k}$. Then, specializing to a spatially homogeneous system, we indeed obtain $|u_k|^2 - |v_{-k}|^2 = (N_c - \tilde{N}_k - 1)/N$, which tends to 1 for $N_c \gg \tilde{N}_k \forall k$ and $N_c \gg 1$. The condition for $u$ and $v$ reported in the text, on the other hand, holds for any value of the density, according to the commutation rules for operators $\hat{a}$ and $\hat{p}_k$. In our formalism we use it to write the two-particle correlation in compact form in Eq. (31).