Non-Equilibrium Bose–Einstein Condensation in a Dissipative Environment

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Abstract:
Solid state quantum condensates can differ from other condensates, such as Helium, ultra-cold atomic gases, and superconductors, in that the condensing quasiparticles have relatively short lifetimes, and so, as for lasers, external pumping is required to maintain a steady state. In this chapter we present a non-equilibrium path integral approach to condensation in a dissipative environment and apply it to microcavity polaritons, driven out of equilibrium by coupling to multiple baths, describing pumping and decay. Using this, we discuss the relation between non-equilibrium polariton condensation, lasing, and equilibrium condensation.

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1   Introduction

The idea of Bose–Einstein condensation of quasiparticles in solid-state structures has a long history, since the early proposals [1–3] that excitons might form a condensate. In more recent years, this has led to work on a variety of systems: excitons in coupled quantum wells [4–7]; excitons in quantum-Hall bilayers [8]; magnons, both in thermal equilibrium [9] and parametrically pumped magnetic insulators [10] as well as within superfluid ^{3}He[11, 12]; and microcavity exciton-polaritons. (For extensive references to experiments see the review [13]). In almost all these cases, the condensate is, to a greater or lesser extent, a non-equilibrium steady state, with pumping compensating for the finite lifetime of the quasiparticles, leading to a flux of particles through the system. Thus, a general question arises: can Bose–Einstein condensation be realised in a strongly dissipative environment, and if so how would it relate to and differ from equilibrium BEC and the laser?

To address these questions, this chapter discusses a field theoretical approach to modelling quantum condensates that are driven out of equilibrium by a flux of particles through the system. We illustrate the technique using an example of microcavity polaritons. Their part-light nature leads to a rather short lifetime that may nonetheless be long enough to have polaritons as well-defined quasiparticles (i.e. strong coupling). Their short lifetimes however lead to an important role of non-equilibrium physics. This naturally provokes questions about the relation to lasing, which occurs in pumped cavities in the weak coupling limit. We consider a polariton system coupled to baths which model pumping and decay processes. Since these baths are not in chemical equilibrium with each other, they drive a flux of particles. The Hamiltonian we use will describe both a laser (if pumped at high temperatures, as discussed below), and Bose condensation if treated in thermal equilibrium, as well as the smooth transition between them. As such, the system of microcavity polaritons provides a particularly rich playground for studying coherence in a dissipative environment, and exploring the differences and similarities between condensates and lasers.

2   Methodology: Modelling the Non-Equilibrium System

For a non-equilibrium system, the density of states and its occupation must both be determined explicitly, as the occupation may be non-thermal. This means that to describe the system fully, one needs at least two Green's functions. We choose here to work with the retarded and Keldysh Green's functions: $D^R(\mathbf{r}, \mathbf{r}', t, t') = -i\theta(t)\langle[\hat{\psi}(\mathbf{r}, t), \hat{\psi}^\dagger(\mathbf{r}', t')]_-\rangle$, $D^K(\mathbf{r}, \mathbf{r}', t, t') = -i\langle[\hat{\psi}(\mathbf{r}, t), \hat{\psi}^\dagger(\mathbf{r}', t')]_+\rangle$, where $[\hat{\psi}, \hat{\psi}^\dagger]_\mp$ is the commutator (anti-commutator). The retarded Green's function describes the response following some applied perturbation. In the frequency domain, $\rho(\mathbf{p}, \omega) = 2\,\mathrm{Im}[D^R(\mathbf{p}, \omega)]$ gives the density of states, while the Keldysh Green's function $D^K(\mathbf{p}, \omega) = -i[2n(\omega) + 1]\rho(\mathbf{p}, \omega)$ accounts for occupation $n(\omega)$.

To determine these Green's functions, we will use a path integral approach[14], discussed further below. Path integrals naturally allow computation of time-ordered correlation functions; in order to instead find the retarded and Keldysh Green's functions we must use the Keldysh contour $\mathcal{C}$. Points on this contour are labelled by $(t, \{+, -\})$, where $+, -$ distinguishes the forward($+$) and backward($-$) branches. The path integral approach will then give contour ordered correlations, denoted by $T_\mathcal{C}$, where fields on the $+$ contour always precede those on the $-$ contour, and fields on the $-$ appear in time reversed order. Then introducing symmetric (classical) and anti-symmetric (quantum) combinations of these fields $\psi_{\mathrm{cl,q}} = [\psi(+, t) \pm \psi(-, t)]/\sqrt{2}$, the Green's functions are given by:

$$D = \begin{pmatrix} D^K & D^R \\ D^A & 0 \end{pmatrix} = -i\left\langle T_\mathcal{C} \begin{pmatrix} \psi_{\mathrm{cl}}(\mathbf{r}, t) \\ \psi_{\mathrm{q}}(\mathbf{r}, t) \end{pmatrix} \left(\psi_{\mathrm{cl}}^\dagger(\mathbf{r}', 0), \psi_{\mathrm{q}}^\dagger(\mathbf{r}', 0)\right)\right\rangle. \tag{1}$$

(D^A is the advanced Green's function, the Hermitian conjugate of D^R). As discussed below, the action in the path integral involves the inverse Green's function: $D^{-1} = \left[\begin{pmatrix} D^K & D^R \\ D^A & 0 \end{pmatrix} \right]^{-1} = \begin{pmatrix} 0 & [D^A]^{-1} \\ [D^R]^{-1} & [D^{-1}]^K \end{pmatrix}$, where $[D^{-1}]^K = -[D^R]^{-1} D^K [D^A]^{-1}$. As an illustration, for a free field $[D_0^R]^{-1} = \hbar\omega - \hbar\omega_p + i\delta$ and $[D_0^{-1}]^K = 2i\delta[2n_B(\hbar\omega) + 1]$, where δ is infinitesimal. The above is for bosonic fields; the results for fermionic fields are similar, but commutators and anti-commutators are interchanged in the definitions of Keldysh and retarded Green's functions.

Polariton System Hamiltonian, and Coupling to Baths

To describe the polariton system we use a model of disorder localised excitons strongly coupled to cavity photons[15–17]. Exciton-exciton interactions are included in this model by allowing only zero or one excitons on a given site, thus describing hard-core bosons. This model has several advantages for our aims: Firstly, this same Hamiltonian has been used to model lasers [18], allowing us to relate polariton condensation to lasing. Secondly, it is known[16] that, in equilibrium, except at extremely low densities, mean-field theory captures the phase diagram of this model rather well. Finally, it allows one to account straightforwardly for exciton nonlinearity within the non-equilibrium mean-field theory.

To describe a hard-core boson, we introduce two fermionic operators $\hat{d}_j^\dagger, \hat{c}_j^\dagger$ that create states representing the presence or absence of an exciton. The operator $\hat{d}_j^\dagger \hat{c}_j$ thus creates an exciton. In this notation, the system Hamiltonian is $\hat{H}_{\text{sys}} = \sum_j \epsilon_j (\hat{d}_j^\dagger \hat{d}_j - \hat{c}_j^\dagger \hat{c}_j) + \sum_{\mathbf{p}} \hbar\omega_{\mathbf{p}} \hat{\Psi}_{\mathbf{p}}^\dagger \hat{\Psi}_{\mathbf{p}} + \sum_{j,\mathbf{p}} g_j (\hat{\Psi}_{\mathbf{p}}^\dagger \hat{c}_j^\dagger \hat{d}_j + \text{H.c.})$. Here ϵ_j is the exciton state energy and g_j is the coupling to photons. The cavity photon dispersion is $\hbar\omega_{\mathbf{p}} = \hbar\omega_0 + \hbar^2 p^2 / 2m_{\text{phot}}$.

The system is driven out of equilibrium by its coupling to separate pumping and decay baths, so that the full Hamiltonian is given by $\hat{H} = \hat{H}_{\text{sys}} + \hat{H}_{\text{bath}}^{\text{pump}} + \hat{H}_{\text{bath}}^{\text{decay}}$. The contribution of the pumping bath is $\hat{H}_{\text{bath}}^{\text{pump}} = \sum_{j,n} \Gamma_{j,n} (\hat{c}_j^\dagger \hat{C}_{j,n} + \hat{d}_j^\dagger \hat{D}_{j,n} + \text{H.c.}) + \sum_{j,n} \nu_{j,n}^\Gamma (\hat{D}_{j,n}^\dagger \hat{D}_{j,n} - \hat{C}_{j,n}^\dagger \hat{C}_{j,n})$. The fermionic operators $\hat{D}_{j,n}^\dagger, \hat{C}_{j,n}^\dagger$ describe the pumping bath modes, and $\Gamma_{j,n}$ is the coupling strength. Similarly, the contribution of the decay bath is $\hat{H}_{\text{bath}}^{\text{decay}} = \sum_{\mathbf{p},p_z} \zeta_{\mathbf{p},p_z} (\hat{\Psi}_{\mathbf{p}}^\dagger \hat{\Xi}_{\mathbf{p},p_z} + \text{H.c.}) + \sum_{\mathbf{p},p_z} \hbar\omega_{\mathbf{p},p_z}^\zeta \hat{\Xi}_{\mathbf{p},p_z}^\dagger \hat{\Xi}_{\mathbf{p},p_z}$, with $\hat{\Xi}_{\mathbf{p},p_z}^\dagger$ describing bulk photon modes. Each confined photon mode $\mathbf{p}$ couples to a separate set of bulk photon modes with various values p_z, corresponding to conservation of in-plane momentum in the coupling between cavity and bulk photon modes.

Path-Integral Formulation

Following Ref. [14], we construct the non-equilibrium generating functional $\mathcal{Z}$ as a coherent state path integral over fields[1] defined on the closed-time-path contour, $\mathcal{C}$. For conciseness, we arrange the fermionic fields into a Nambu vector $\Lambda = (d, c)^{\text{T}}$. Formally, the partition function is thus: $\mathcal{Z} = \int \prod_{\mathbf{p},p_z,j,n} \mathcal{D}[\Psi_{\mathbf{p}}, \Lambda_j, C_{j,n}, D_{j,n}, \Xi_{\mathbf{p},p_z}] e^{iS}$, where the action[2] $S = \int_{\mathcal{C}} dt \langle \Psi_{\mathbf{p}}(t) \Lambda_j(t) C_{j,n}(t) D_{j,n}(t) \Xi_{\mathbf{p}pz}(t) | i\hbar\partial_t - H | \Psi_{\mathbf{p}}(t) \Lambda_j(t) C_{j,n}(t) D_{j,n}(t) \Xi_{\mathbf{p}pz}(t) \rangle$, in terms of coherent states specified by the fields $\Psi_{\mathbf{p}}, \Lambda_j, C_{j,n}, D_{j,n}, \Xi_{\mathbf{p},p_z}$ on the Keldysh time contour $\mathcal{C}$. In what follows, we consider fields in the classical and quantum rather than forward and backward basis.

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[1] In keeping with the convention of Ref. [14], we also refer to field amplitudes defined at discrete momenta, such as $\Psi_{\mathbf{p}}$, as fields. We note that $\mathcal{Z}$ is necessarily a functional integral, as we must account for a continuum of paths taken by Ψ, Λ, C, D, Ξ (and their complex conjugates) as functions of the continuous time variable t.

[2] When evaluating things we tend to take the continuum limit over $\mathbf{p}$, making the partial time-derivative more convenient and appropriate [14].

Treatment of Environment

For the bath Hamiltonian given above, the action S contains only terms linear or quadratic in the bath fields $C_{j,n}, D_{j,n}, \Xi_{\mathbf{p},p_z}$ and their conjugates. Thus, the integral over these fields is Gaussian, and can straightforwardly be evaluated analytically. For the decay bath one thus finds:

$$S_{\text{bath}}^{\text{decay}} = -\iint_{-\infty}^{\infty} dt dt' \sum_{\mathbf{p}} \Psi_{\mathbf{p}}^*(t) \sigma_1^{\text{K}} \sum_{p_z} \zeta_{\mathbf{p},p_z}^2 \left[(i\hbar\partial_t - \hbar\omega_{\mathbf{p},p_z}^\zeta)\sigma_1^{\text{K}}\right]^{-1} \sigma_1^{\text{K}} \Psi_{\mathbf{p}}(t'), \qquad (2)$$

where σ_1^{K} is a Pauli matrix in the (Keldysh) space of quantum and classical fields. By definition, the bath has a large number of modes, and these modes thermalise rapidly compared to system-bath interactions. Hence we may take the bath occupation functions as fixed, and then allow the system distribution to be self-consistently determined. The Green's function of a free bosonic field is $[(i\hbar\partial_t - \hbar\omega_{\mathbf{p}}^\zeta)\sigma_1^{\text{K}}]^{-1} = \begin{pmatrix} D_{\mathbf{p}}^K(t-t') & D_{\mathbf{p}}^R(t-t') \\ D_{\mathbf{p}}^A(t-t') & 0 \end{pmatrix}$, where we have written $\mathbf{p} = (\mathbf{p}, p_z)$. In frequency space the retarded and Keldysh Green's functions are given by $D_{\mathbf{p}}^R(\omega) = [\hbar\omega - \hbar\omega_{\mathbf{p}}^\zeta + i\delta]^{-1}, D_{\mathbf{p}}^K(\omega) = (-2\pi i)F_\Xi(\omega)\delta(\omega - \omega_{\mathbf{p}}^\zeta)$. Here $F_\Xi(E) = 1 + 2n_\Xi(E)$ where $n_\Xi(E)$ is the occupation function for the bath modes, which can have any form. For our situation, the decay bath is empty.

Before proceeding further, we make some simplifying assumptions about the baths. We assume the bath frequencies $\omega_{\mathbf{p},p_z}^\zeta$ form a dense spectrum, and the coupling constant $\zeta_{\mathbf{p},p_z} = \zeta(\omega_{\mathbf{p},p_z}^\zeta)$ is a smooth function. We may then replace summation over bath modes by integration. Then, taking the bath density of states and $\zeta(\omega)$ to be frequency independent, we have:

$$S_{\text{bath}}^{\text{decay}} = \int_{-\infty}^{\infty} d\omega \sum_{\mathbf{p}} \Psi_{\mathbf{p}}^*(\omega) \begin{pmatrix} 0 & -i\kappa_c \\ i\kappa_c & 2i\kappa_c F_\Xi(\hbar\omega) \end{pmatrix} \Psi_{\mathbf{p}}(\omega). \qquad (3)$$

We follow an analogous procedure for the pumping baths (see [19] for details).

Integration over Fermionic Fields

After integrating over the bath degrees of freedom the full action S is:

$$S = \iint_{-\infty}^{\infty} dt dt' \left[\sum_j \Lambda_j^*(t) G_j^{-1}(t,t') \Lambda_j(t') + \sum_{\mathbf{p}} \Psi_{\mathbf{p}}^*(t) D_{(0),\mathbf{p}}^{-1}(t,t') \Psi_{\mathbf{p}}(t')\right],$$

$$\text{where } D_{(0),\mathbf{p}}^{-1}(t,t') = \begin{pmatrix} 0 & i\hbar\partial_t - \hbar\omega_{\mathbf{p}} - i\kappa_c \\ i\hbar\partial_t - \hbar\omega_{\mathbf{p}} + i\kappa_c & 2i\kappa_c F_\Xi(t - t') \end{pmatrix}. \quad (4)$$

To specify the exciton Green's function G_j we first introduce the abbreviations $\lambda_{\text{cl}}(t) = \sum_{\mathbf{p}} g_j \Psi_{\mathbf{p},cl}(t)/\sqrt{2}$ and $\lambda_{\text{q}}(t) = \sum_{\mathbf{p}} g_j \Psi_{\mathbf{p},q}(t)/\sqrt{2}$ so that:

$$G_j^{-1} = \begin{pmatrix} 0 & -\lambda_{\text{q}}(t) & i\hbar\partial_t - \epsilon_j - i\gamma_x & -\lambda_{\text{cl}}(t) \\ -\lambda_{\text{q}}^*(t) & 0 & -\lambda_{\text{cl}}^*(t) & i\hbar\partial_t + \epsilon_j - i\gamma_x \\ i\hbar\partial_t - \epsilon_j + i\gamma_x & -\lambda_{\text{cl}}(t) & 2i\gamma_x F_D(i\hbar\partial_t) & -\lambda_{\text{q}}(t) \\ -\lambda_{\text{cl}}^*(t) & i\hbar\partial_t + \epsilon_j + i\gamma_x & -\lambda_{\text{q}}^*(t) & 2i\gamma_x F_C(i\hbar\partial_t) \end{pmatrix}, \qquad (5)$$

where $F_{C,D}(E) = 1 - 2n_{C,D}(E)$ with $n_{C,D}(E)$ the pumping bath occupation functions. As the occupation functions of all baths appear in this action, they compete to set the occupation function of the polaritons. This non-equilibrium action thus combines strong exciton-photon coupling with the effects of dissipation due to the open nature of the system. The action is

quadratic also in the fermionic fields Λ_j, so we can also integrate over these fields to get the effective action for the photon field alone:

$$S = -i \sum_j \mathrm{Tr}\left\{\ln G_j^{-1}\right\} + \iint_{-\infty}^{\infty} dt dt' \sum_{\mathbf{p}} \Psi_{\mathbf{p}}^*(t) D_{(0),\mathbf{p}}^{-1}(t,t') \Psi_{\mathbf{p}}(t'). \tag{6}$$

As yet, we have made no assumption about what form $\Psi_{\mathbf{p}}(t)$ takes, however, since $\mathrm{Tr}\{\ln G_j^{-1}\}$ involves $\Psi_{\mathbf{p}}(t)$, this effective action is nonlinear, so to proceed further analytically, some expansion or approximation scheme is required. Section 3 therefore discusses the mean-field theory of this model, and how it relates to laser theory as well as equilibrium results.

3   Mean-Field Condition for a Coherent State

The mean-field theory of the non-equilibrium system describes a self-consistent steady state, which may be found by evaluating the saddle point of S with respect to photon field, $\delta S / \delta \Psi_{\mathbf{p},cl}^* = \delta S / \delta \Psi_{\mathbf{p},q}^* = 0$. The first equation is satisfied if the quantum component vanishes, $\Psi_{\mathbf{p},q} = 0$. For the classical component, we write $\Psi_{\mathbf{p},cl} = \sqrt{2}\phi_{\mathbf{p}}$, so $\phi_{\mathbf{p}}$ corresponds to the expectation of photon annihilation. If condensed, we consider the ansatz $\phi_{\mathbf{p}} = \phi_0 \exp(-i\mu_S t)\delta_{\mathbf{p},0}$, controlled by the parameters ϕ_0, μ_S. For this ansatz to satisfy the saddle point equation, one requires

$$(\hbar\omega_0 - \mu_S - i\kappa_c)\phi_0 = \frac{i}{2} \int \frac{d\nu}{2\pi} G_{c_j^\dagger d_j}^K(\nu). \tag{7}$$

Putting the $G_{c^\dagger d}^K$ component of Eq. (5) into Eq. (7) and defining $E_j^2 = (\epsilon_j - \mu_s/2)^2 + g_j^2 \phi_0^2$ we have the saddle point (mean-field) equation:

$$(\hbar\omega_0 - \mu_S - i\kappa_c)\phi_0 = \sum_j g_j^2 \phi_0 \gamma_x$$

$$\times \int \frac{d\nu}{2\pi} \frac{[F_D(\nu) + F_C(\nu)]\nu + [F_D(\nu) - F_C(\nu)](\epsilon_j - \mu_S/2 + i\gamma_x)}{[(\nu - E_j)^2 + \gamma_x^2][(\nu + E_j)^2 + \gamma_x^2]}. \tag{8}$$

As noted above, the pumping bath occupations are imposed by choice, and we choose these to model a thermalised reservoir of high energy excitons, with a population set by the strength of pumping. In order to obey on average the constraint that we consider two-level systems, we take $n_C(\nu) + n_D(\nu) = 1$. Introducing parameters μ_B, β_B to describe the occupation and temperature of this exciton reservoir, we thus define:

$$F_{C,D}(\nu) = \tanh\left[\frac{\beta_B}{2}\left(\nu \pm \frac{\mu_B - \mu_S}{2}\right)\right]. \tag{9}$$

(μ_S appears here via a gauge transform required to remove explicit time dependence from the effective action). If there were no exciton-photon coupling the excitonic two-level systems would be thermally occupied, i.e. $\langle d_j^\dagger d_j - c_j^\dagger c_j \rangle = -\tanh[\beta_B(\epsilon_j - \mu_B/2 + \mu_S/2)/2]$.

As anticipated above, Eq. (8) is rather general, encompassing limits that correspond both to the equilibrium gap-equation for our model polariton system (discussed in Section 3), as well as being capable of recovering the standard laser limit (discussed in Section 3). In addition, if one extends this approach to slowly varying condensates, then as discussed in Section 3, one may make contact with the complex Gross–Pitaevskii approach.

Equilibrium Limit of Mean-Field Theory

The simplest limit of the self-consistency equation, Eq. (8), is the thermal equilibrium limit, which corresponds to taking $\gamma_x, \kappa_c \to 0$. In taking this limit, it is necessary to send $\kappa_c \to 0$ first and then $\gamma_x \to 0$. This is because the self-consistency equation contained only the coupling of coherent photons to the decay bath, hence the decay bath cannot impose a non-trivial distribution on the system, while the pumping bath can. In order to satisfy Eq. (8) with $\kappa_c = 0$, the imaginary part of the right hand side must vanish. The most general way to achieve this is to set $F_D(\nu) = F_C(\nu)$, which, considering Eq. (9), implies $\mu_S = \mu_B$. That is, in the absence of decay, one has chemical equilibrium between the pumping bath and the system. With $\mu_S = \mu_B$, the remaining part of Eq. (8) becomes:

$$(\hbar\omega_0 - \mu_B)\phi_0 = \sum_j g_j^2 \phi_0 \gamma_x \int \frac{d\nu}{2\pi} \frac{2\tanh(\beta_B\nu/2)\,\nu}{[(\nu - E_j)^2 + \gamma_x^2][(\nu + E_j)^2 + \gamma_x^2]}. \tag{10}$$

In the limit of small γ_x, one may use that $\lim_{\gamma_x \to 0} 2\gamma_x/[(\nu - E_j)^2 + \gamma_x^2] = 2\pi\delta(\nu - E_j)$ to find $(\hbar\omega_0 - \mu_B)\phi_0 = \sum_j (g_j^2 \phi_0/2E_j) \tanh(\beta_B E_j/2)$. This is the equilibrium mean-field theory [15–17] of the system Hamiltonian introduced above.

High Temperature Limit of Mean-Field Theory — Laser

An alternative limit to thermal equilibrium is that of a simple laser. This limit too can be recovered from Eq. (8), in this case by taking $F_{C,D}(\nu)$ to be frequency independent. This frequency independence can be recovered from Eq. (9) in the limit $T \to \infty$, while keeping $\mu_B \propto T$ in order that the bath population remains fixed. Another interpretation of this is that infinite temperature corresponds to white noise, i.e. a Markovian approximation, where the occupation of the bath modes is frequency independent. [In contrast, Eq. (8) has a flat density of states of the bath, but a non-Markovian, i.e. frequency dependent, occupation].

Taking $F_{C,D}(\nu)$ to be frequency independent, the integral in Eq. (8) can then be simply evaluated by contour integration to give:

$$(\hbar\omega_0 - \mu_S - i\kappa_c)\phi_0 = \sum_j g_j^2 \phi_0 (F_D - F_C) \frac{\epsilon_j - \mu_S/2 + i\gamma_x}{4(E_j^2 + \gamma_x^2)}. \tag{11}$$

The term on the right hand side, describing the two-level system polarisation, is proportional to the bath inversion $N_0 = (n_D - n_C) = -(F_D - F_C)/2$. In the limit $\phi_0 \to 0$, this equation recovers the standard threshold condition for a laser [18]. This is clear if one restricts to $g_j = g, \epsilon_j = \epsilon$ so the sum is replaced by a factor n, and one assumes resonance, $2\epsilon = \hbar\omega_0 = \mu$, which yields: $2\kappa_c\gamma_x/g^2 = nN_0 =$ total inversion.

Low Density Limit: Complex Gross–Pitaevskii Equation

Equation (8) is written for a uniform steady state, but in many cases, it is interesting to allow for solutions that vary slowly in time and space. To do this rigorously requires some care, but the basic idea can be described simply: One may start by writing Eq. (8) in the form $(\mu_S + i\kappa_c - \hbar\omega_0)\phi = \chi[\phi, \mu_S]\phi$, where $\chi[\phi, \mu_S]$ is a nonlinear complex susceptibility. If one then separates the fast and slow time dependence $\Psi(\mathbf{r}, t) = \phi(\mathbf{r}, t)e^{-i\omega_0 t}$, one may write $(i\hbar\partial_t + i\kappa_c - [V(\mathbf{r}) - \hbar^2\nabla^2/2m])\phi(\mathbf{r}, t) = \chi[\phi(\mathbf{r}, t)]\phi(\mathbf{r}, t)$, having introduced an external potential $V(\mathbf{r})$. Then, by making a gradient and Taylor expansion of the nonlinear complex susceptibility $\chi[\phi]$, one is naturally led to a complex Gross–Pitaevskii equation: $i\hbar\partial_t\phi = (-\hbar^2\nabla^2/2m^* + V_{\text{eff}}(\mathbf{r}) + U|\phi|^2 + i[\gamma_{\text{net}}(\mu_B) - \Gamma|\phi|^2])\phi$, where Γ represents a nonlinearity of the imaginary part of the susceptibility. The dynamics of the excitons are responsible for producing an effective polariton

mass and effective potential. In some cases, it may also be important to consider the dynamics of the reservoir excitons more carefully, by introducing an extra degree of freedom to describe them.

4   Applications: Fluctuations and Instability Towards BEC

So far, we have discussed only the mean-field properties of the non-equilibrium polariton condensate. We next consider fluctuations about this mean field, and in particular the photon Green's function. This is important for several reasons. Firstly, knowledge of the fluctuations determines whether a state is stable (i.e. do fluctuations grow or decay in time). Secondly, the photon Green's function describes the fluctuation contribution to physical observables such as luminescence and absorption spectra.

To determine the photon Green's function, one may start from Eq. (6), and expand $\Psi = \phi + \psi$ to second order in ψ. The inverse photon Green's function has two parts, one from the bare photon action [given in Eq. (4)], and one from expanding the trace over excitons. To determine the exciton part one may write $G_j^{-1} = (G_j^{\mathrm{sp}})^{-1} + \delta G_j^{-1}$, where G_j^{sp} is the fermionic Green's function including the mean-field photon field ϕ, and δG_j^{-1} is the photon fluctuation part given by: $\delta G_j^{-1} = -g_j(\psi_{\mathrm{q}}^* \sigma_-^{dc} + \psi_{\mathrm{q}} \sigma_+^{dc})\sigma_0^{\mathrm{K}}/\sqrt{2} - g_j(\psi_{\mathrm{cl}}^* \sigma_-^{dc} + \psi_{\mathrm{cl}} \sigma_+^{dc})\sigma_1^{\mathrm{K}}/\sqrt{2}$ where σ^{dc} are Pauli matrices in the space of fermionic fields c, d. The action then depends on

$$\mathrm{Tr}\left\{\ln G_j^{-1}\right\} = \mathrm{Tr}\left\{\ln\left[(G_j^{\mathrm{sp}})^{-1}\right] + G_j^{\mathrm{sp}}\delta G_j^{-1} - \frac{1}{2}G_j^{\mathrm{sp}}\delta G_j^{-1}G_j^{\mathrm{sp}}\delta G_j^{-1}\right\}. \tag{12}$$

The last term gives a contribution quadratic in ψ, which contributes to the inverse photon Green's function.

When considering the condensed state, it is necessary to allow for anomalous correlations. This requires writing the Green's function in a (Nambu) vector space of (ψ_k, ψ_{-k}^*), combined with the $\pm$ space due to the Keldysh/retarded/advanced structure. Thus, in the condensed case, the Green's function is a 4×4 matrix, while when non-condensed it is only a 2×2 matrix. We begin by considering fluctuations in the normal state, and the nature of the instability to the condensate, and then in Section 4 briefly discuss fluctuations in the condensed state.

Normal State Green's Functions and BEC Instability

In the normal state, the spectrum and its occupation are determined by three real functions, the real and imaginary parts of the inverse retarded Green's function $[D^R(\mathbf{p},\omega)]^{-1} = A(\mathbf{p},\omega) + iB(\omega)$ and the inverse Keldysh Green's function $[D^{-1}(\omega)]^K = iC(\omega)$. These functions can be read off from the fluctuation action. Once these functions are known, one may invert these expressions to find the retarded and Keldysh Green's functions, and thus determine the density of states $\rho(\mathbf{p},\omega) = -2\,\mathrm{Im}[D^R(\mathbf{p},\omega)]$ and occupation of the modes $2n_\psi(\omega) + 1 = iD^K(\omega)/\rho(\omega)$:

$$\rho(\mathbf{p},\omega) = \frac{2B(\omega)}{A(\mathbf{p},\omega)^2 + B(\omega)^2}, \qquad n_\psi(\omega) = \frac{1}{2}\left[\frac{C(\omega)}{2B(\omega)} - 1\right]. \tag{13}$$

In terms of these, physical observables can be found, such as the luminescence $\mathcal{L}(\mathbf{p},\omega) = \rho(\mathbf{p},\omega)n_\psi(\omega)$. The roles of $A(\mathbf{p},\omega), B(\omega), C(\omega)$ can be understood by considering the contribution from the bare photon action. In this case $A(\mathbf{p},\omega) = \hbar\omega - \hbar\omega_p$ determines the locations of the normal modes, $B(\omega) = \kappa_c$ gives the linewidth of these modes and $C(\omega) = 2\kappa_c[2n_\Xi(\omega) + 1]$ describes their occupation. Including the effect of the excitons, the zeros of $A(\mathbf{p},\omega)$ now describe polaritons, rather than bare photons, and in addition $B(\omega)$ is no longer constant, hence it plays a second role: If $B(\omega)$ vanishes at some $\hbar\omega = \mu_{\mathrm{eff}}$ then this causes the occupation $n_\psi(\mu_{\mathrm{eff}})$ to
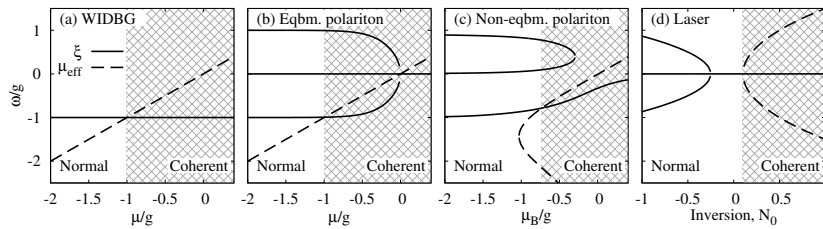
Figure 1: Trajectories of zeros of $A(\omega, \mathbf{p} = 0), B(\omega)$ (i.e. normal modes ξ_0 and effective chemical potential μ_{eff}) for: (a) equilibrium weakly interacting dilute Bose gas (WIDBG); (b) equilibrium polariton condensate; (c) non-equilibrium polariton condensate; (d) Maxwell-Bloch laser.

diverge. However, as long as $A(\mathbf{p}, \mu_{\text{eff}}/\hbar)$ does not vanish, the density of states will be zero at μ_{eff}, so the luminescence will remain finite.

A diverging occupation and vanishing density of states is exactly what would, in equilibrium, occur at the chemical potential, hence the identification μ_{eff}. Although the non-equilibrium system may be far from thermal, the emergence of zeros of $B(\omega)$ thus still describes an effective chemical potential. To ensure $\forall_{\mathbf{p}} A(\mathbf{p}, \mu_{\text{eff}}/\hbar) \neq 0$, it is necessary that μ_{eff} is below any of the zeros of $A(\mathbf{p}, \omega)$, i.e. the chemical potential is below all the polariton modes, as expected in the normal state. We next discuss the instability as μ_{eff} approaches the bottom of the polariton spectrum. If μ_{eff} is near the bottom of the polariton spectrum, we can expand $A(\mathbf{p}, \omega), B(\omega)$ near their simultaneous zero, i.e. $A(\mathbf{p}, \omega) = \alpha(\hbar\omega - \xi_{\mathbf{p}})$, and $B(\omega) = \beta(\hbar\omega - \mu_{\text{eff}})$. One may then find where the actual complex poles $\omega_{\mathbf{p}}^{(D^R)}$ of the retarded Green's function occur: $\hbar\omega_{\mathbf{p}}^{(D^R)} = [(\alpha^2\xi_{\mathbf{p}} + \beta^2\mu_{\text{eff}}) + i\alpha\beta(\mu_{\text{eff}} - \xi_{\mathbf{p}})][\alpha^2 + \beta^2]^{-1}$. These poles determine the response to a small perturbation; thus, for perturbations to decay the poles must have a negative imaginary part. Hence, if $\mu_{\text{eff}} > \xi_{\mathbf{p}}$, then perturbations at that $\mathbf{p}$ grow and the normal state is unstable. One may also show that the point where $\mu_{\text{eff}} = \xi_0$, coincides with the first point where it is possible to satisfy the mean-field equation, Eq. (8) with $\phi = 0$ and $\mu_S = \mu_{\text{eff}} = \xi_0$.

One can now understand the behaviour of the non-equilibrium system as pumping (and hence μ_B) increases: At very weak pumping, where μ_B is large and negative, $B(\omega)$ is always positive (i.e. decay dominates over gain), and no μ_{eff} exists. As μ_B increases, a region of negative $B(\omega)$ develops, and the boundaries of this region define μ_{eff} as discussed above. As long as $\mu_{\text{eff}} < \xi_0$ the normal state remains stable. At the critical pumping power, μ_{eff} then reaches the lower polariton mode at $\mathbf{p} = 0$, the normal state becomes marginally stable, and the mean-field equation can be satisfied. Beyond this point, the normal state would be unstable, but the condensed solution is now possible (and can be shown to be stable).

Fig. 1(c) shows the evolution of μ_{eff} and ξ for the non-equilibrium polariton condensate. For comparison, Fig. 1(b) shows the behaviour of the system Hamiltonian in thermal equilibrium and Fig. 1(a) that of weakly interacting and dilute Bose gas. One may note that despite the absence of a thermal distribution in Fig. 1(c), the scenarios of normal state instability in these two figures are very similar, and would remain the same as long as the distribution function develops a divergence while the polariton system remains in strong coupling. In the next section, we discuss a case where the instability is somewhat different, that of the simple laser discussed in Section 3 and shown in Fig. 1(d).

Normal-State Instability for a Simple Laser

As in Section 3, one may contrast the behaviour of the non-equilibrium condensate to that of a simple laser described by the Maxwell-Bloch equations [18], which corresponds to the high tem-

perature (white noise) limit of our problem. As the above analysis concerns the retarded Green's function, we must define this function for the Maxwell-Bloch equations. The retarded Green's function describes the linear response of the system to an applied field, and so if one introduces a field $Fe^{-i\omega t}$ coupled to the photon field, then one has by definition $\psi(t) = iD^R(\omega)Fe^{-i\omega t}$. One then finds:

$$[D^R(\omega)]^{-1} = A(\omega) + iB(\omega) = \hbar\omega - \hbar\omega_0 + i\kappa_c + \sum_j \frac{g_j^2 N_0}{\hbar\omega - 2\epsilon_j + i2\gamma_x}. \qquad (14)$$

For this form, $B(\omega)$ can only become negative if N_0 is sufficiently large. Restricting as in Section 3 to $g_j = g, \epsilon_j = \epsilon = \hbar\omega_0/2$ one finds the requirement for gain is $g^2 n N_0 > 2\kappa_c\gamma_x$, which is again the laser threshold condition. In this same restricted case, the zeros of $A(\omega)$ behave as follows: A solution $\xi = 0$ always exists, and if $N_0 < -4\gamma_x^2/g^2 n$, an extra pair of roots exist. The evolution of these zeros is shown in Fig. 1(d). One may note that for the Maxwell-Bloch equations, strong-coupling (i.e. splitting of the modes ξ) collapses before condensation (lasing) occurs, while for the polariton condensate, condensation occurs while still strongly coupled.

Fluctuations of the Condensed System

As noted earlier, if condensed, the Green's function is a 4×4 matrix, so the derivation of the spectrum becomes more complicated, however the essential features can be explained by general arguments. The following discussion is thus based on symmetry arguments. (The full derivation matches these results [19]). The form of the inverse retarded Green's function is constrained by the following requirements: there must be symmetry under $\mathbf{p} \to -\mathbf{p}$; the modes must in general have a finite linewidth; however at $\mathbf{p} \to 0$, there must be a mode with vanishing frequency and vanishing linewidth corresponding to global phase rotations of the condensate. These three considerations determine the leading order behaviour of $D^R(\mathbf{p}, \omega)$ for small $\omega, \mathbf{p}$. Using these ideas, one may then write:

$$D^R(\mathbf{p}, \omega) = \frac{C}{\det([D^R]^{-1})} = \frac{C}{\omega^2 + 2i\omega x - c^2 p^2}. \qquad (15)$$

The parameters x, c describe the linewidth and sound velocity.

¿From this form of $D^R(\mathbf{p}, \omega)$, one finds the poles are given by $\omega_{\mathbf{p}}^{(D^R)} = -ix \pm i\sqrt{x^2 - c^2 p^2}$. At long wavelengths, these are diffusive (only an imaginary part exists), and only above a critical momentum does a real part emerge. Given the generality of the argument leading to this result, it is unsurprising to find the same structure emerges from other approaches, see e.g. [20]. Similar results also occur for the case of a parametrically pumped polariton system [21] . The absence of a linear dispersion of energy vs momentum in the condensed state affects some aspects of superfluidity in this non-equilibrium system, however there are also aspects of superfluid behaviour that survive[22, 23]

5   Connection to Other Approaches

The language of the Keldysh path integral, and the Keldysh Green's functions provide a natural bridge to many other approaches that have been used to model non-equilibrium polariton condensation. We have already discussed above the connection between the mean-field theory, i.e. the saddle-point of the Keldysh action, and the complex Gross–Pitaevskii equation [20, 24, 25]. In order to go beyond mean-field theory, the approach discussed in this chapter makes use of Keldysh/retarded/advanced Green's functions to describe both the occupation of a mode, and the density of states. These Green's functions can naturally be related to the one particle density

matrix $\rho(\mathbf{r}, \mathbf{r}', t) = \langle \hat{\psi}^\dagger(\mathbf{r}, t)\hat{\psi}(t, \mathbf{r}')\rangle = i(D^K - D^R + D^A)(\mathbf{r}, \mathbf{r}', t, t)/2$. Direct time evolution of the one particle density matrix has been used to treat polaritons in zero-[26] and one-dimensional geometries [27], and stochastic methods for simulating density matrix evolution have been used to describe a number of properties of polariton condensates, see e.g. [28, 29]. Another stochastic approach used for exciton condensation is the Heisenberg–Langevin equations [30]. Such an approach again connects naturally to the Keldysh formalism, with the Keldysh self energy due to the bath corresponding directly to the noise correlator of the Heisenberg–Langevin approach, and the retarded self energy corresponding to the dissipation term. Finally, there is also a clear connection between the Keldysh Green's functions and the quantum Boltzmann equation (see e.g. [31, 32] for a derivation of the quantum Boltzmann equation from the equations of motion for the Keldysh Green's functions). There have been many works using the Boltzmann equation to model kinetics of polariton condensation [33–40]. By considering how the quantum Boltzmann equation arises from the Keldysh Green's function, one may note that in order to correctly describe the coherence properties of the condensed state, one must include anomalous retarded self energies, modifying the polariton spectrum.

Acknowledgements

Bibliography

[1] L. V. Keldysh and Y. V. Kopaev, Sov. Phys. Solid State **6**, 2219 (1965).

[2] J. M. Blatt, K. W. Böer, and W. Brandt, Phys. Rev. **126**, 1691 (1962).

[3] S. A. Moskalenko, Sov. Phys.: Solid State **4**, 199 (1962).

[4] L. V. Butov, C. W. Lai, A. L. Ivanov, A. C. Gossard, and D. S. Chemla, Nature **417**, 47 (2002).

[5] L. V. Butov, A. C. Gossard, and D. S. Chemla, Nature **418**, 751 (2002).

[6] D. Snoke, S. Denev, Y. Liu, L. Pfeiffer, and K. West, Nature **418**, 754 (2002).

[7] L. V. Butov, J. Phys.: Condens. Matter **16**, R1577 (2004).

[8] J. P. Eisenstein and A. H. MacDonald, Nature **432**, 691 (2004).

[9] C. Rüegg, N. Cavadinin, A. Furrer, H.-U Güdel, K. Krämer, H. Mukta, A. Wildes, K. Habicht, and P. Worderwisch, Nature **423**, 62 (2003).

[10] S. O. Demokritov, V. E. Demidov, O. Dzyapko, G. A. Melkov, A. A. Serga, B. Hillebrands, and A. N. Slavin, Nature **443**, 430 (2006).

[11] G. Volovik, J. Low. Temp. Phys. **153**, 266 (2008).

[12] Y. M. Bunkov and G. V. Volovik, J. Phys.: Condens. Matter **22**, 1 (2010).

[13] H. Deng, H. Haug, and Y. Yamamoto, Rev. Mod. Phys. **82**, 1489 (2010).

[14] A. Kamenev, *Many-body theory of non-equilibrium systems* (Elsevier, Amsterdam, The Netherlands, 2005), p. 177.

[15] P. R. Eastham and P. B. Littlewood, Solid State Commun. **116**, 357 (2000).

[16] J. Keeling, P. R. Eastham, M. H. Szymanska, and P. B. Littlewood, Phys. Rev. Lett. **93**, 226403 (2004).

[17] F. M. Marchetti, J. Keeling, M. H. Szymańska, and P. B. Littlewood, Phys. Rev. Lett. (2006).

[18] H. Haken, *The semiclassical and quantum theory of the laser* (Academic Press, London, 1970), p. 201.

[19] M. H. Szymańska, J. Keeling, and P. B. Littlewood, Phys. Rev. B **75**, 195331 (2007).

[20] M. Wouters and I. Carusotto, Phys. Rev. A **76**, 043807 (2007).

[21] M. Wouters and I. Carusotto, Phys. Rev. Lett. **99**, 140402 (2007).

[22] M. Wouters and I. Carusotto, Phys. Rev. Lett. **105**, 20602 (2010).

[23] J. Keeling, Phys. Rev. Lett. **107**, 080402 (2011).

[24] M. Wouters, I. Carusotto, and C. Ciuti, Phys. Rev. B **77**, 115340 (2008).

[25] J. Keeling and N. G. Berloff, Phys. Rev. Lett. **100**, 250401 (2008).

[26] D. M. Whittaker and P. R. Eastham, Europhys. Lett. **87**, 27002 (2009).

[27] I. G. Savenko, E. B. Magnusson, and I. A. Shelykh, Phys. Rev. B **83**, 165316 (2011).

[28] I. Carusotto and C. Ciuti, Phys. Rev. B **72**, 125335 (2005).

[29] M. Wouters and V. Savona, Phys. Rev. B **79**, 165302 (2009).

[30] B. Mieck and H. Haug, Phys. Rev. B **66**, 075111 (2002).

[31] E. M. Lifshitz and L. P. Pitaevskii, *Statistical Physics, Part II* (Butterworth–Heinemann, Oxford, UK, 1980).

[32] L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (W. A. Benjamin, Menlo Park, CA, USA, 1962).

[33] F. Tassone, C. Piermarocchi, V. Savona, A. Quattropani, and P. Schwendimann, Phys. Rev. B **56**, 7554 (1997).

[34] F. Tassone and Y. Yamamoto, Phys. Rev. B **59**, 10830 (1999).

[35] G. Malpuech, A. Di Carlo, A. Kavokin, J. J. Baumberg, M. Zamfirescu, and P. Lugli, Appl. Phys. Lett. **81**, 412 (2002).

[36] G. Malpuech, A. Kavokin, A. Di Carlo, and J. J. Baumberg, Phys. Rev. B **65**, 153310 (2002).

[37] D. Porras, C. Ciuti, J. J. Baumberg, and C. Tejedor, Phys. Rev. B **66**, 085304 (2002).

[38] T. D. Doan, H. T. Cao, D. B. Tran Thoai, and H. Haug, Phys. Rev. B **72**, 085301 (2005).

[39] T. D. Doan, H. T. Cao, D. B. Tran Thoai, and H. Haug, Phys. Rev. B **74**, 115316 (2006).

[40] T. D. Doan, H. T. Cao, D. B. Tran Thoai, and H. Haug, Phys. Rev. B **78**, 205306 (2008).