Dynamics of an Exciton-Polariton Condensate

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Abstract

Polariton fluids present a unique opportunity to study a strongly interacting, non-equilibrium phase of matter with condensate properties. Under the right circumstances, these systems can acquire superfluid properties, such as dissipationless flow around a defect or topological excitations like vortices and solitons. The low-mass, driven-dissipative nature of the system presents certain advantages to observing these effects, but also introduces extra features that make it differ from a conventional quantum fluid.
1 Introduction

Bose-Einstein Condensates are some of the most impressive examples of quantum mechanics on a macroscopic scale. Unfortunately, they are also some of the most difficult states of matter to observe. Cooling down an atomic condensate requires reaching temperatures on the order of a few Kelvin, or even less. In addition, it is not immediately obvious how to observe parameters like the local phase of the condensate.

It turns out that a certain quasiparticle of quasiparticles can solve both of these problems: the exciton-polariton. Exciton-polaritons, or just polaritons for short, appear inside a special type of semiconductor optical cavity. Under the right conditions, cavity photons can couple to excitons, which themselves are quasiparticles made from electron-hole pairs in the semiconductor. The coupling of all these factors makes the polariton an extremely versatile quasiparticle, with a tunable mass that allows it to condense at much higher temperatures than regular atoms. However, polaritons also have an extremely short lifetime, on the order of a few picoseconds [1]. This means that fluids composed of polaritons are strongly non-equilibrium in nature: the steady state is only achieved when the rate of polariton gain and polariton decay balance out. This has multiple consequences on the system: some are nuisances, others are more interesting, but some make the polariton condensate a unique testing ground for studying superfluid hydrodynamics.

2 Exciton-Polariton Condensates

We will start with the Hamiltonian of a linear optical cavity [1]

\[ H_{\text{cav}} = \int \frac{d^2 k}{(2\pi)^2} \sum_{\sigma} \hbar \omega_{\text{cav}}(k) \hat{a}^\dagger_{\sigma}(k) \hat{a}_{\sigma}(k) \]  

We can immediately notice one major difference with conventional systems: there are only two degrees of freedom for the wavevector \( k \). The \( z \) component of the wavevector, \( k_z \), is forced by the cavity to be quantized in units of \( \pi/l \), where \( l \) is the cavity length [1]. The in-plane components of the wavevector \( k_x \) and \( k_y \) are not fixed by such conditions and, provided they are much less than \( k_z \), have the dispersion relation

\[ \omega_{\text{cav}}(k) \approx \frac{ck_z}{n_0} \left( 1 + \frac{\hbar k^2}{2m_{\text{cav}}} \right) \]

Where

\[ m_{\text{cav}} = \frac{\hbar n_0 k_z}{c} \]

Hence a cavity photon acts like a 2-dimensional particle with an effective mass \( m_{\text{cav}} \).
In nonlinear optical materials where the rotating-wave approximation holds (i.e. photon number is conserved as photon energies exceed other energy scales), 4-photon interactions may also become significant [1]:

$$ \mathcal{H}_{\text{int}} = \frac{1}{2} \int \frac{d^2 k d^2 k' d^2 q}{(2\pi)^6} \sum_{\sigma, \sigma'} V_{\sigma, \sigma'}(k, k', q) \hat{a}_\sigma^\dagger(k + q) \hat{a}_{\sigma'}^\dagger(k') \hat{a}_{\sigma'}(k') \hat{a}_\sigma(k) $$

(5)

### 2.1 Excitons and their Interactions

Excitons are primarily studied in optical quantum wells. These are layered materials about 10nm thick where the conduction bands of the "walls" are higher than that of the center, while the valence bands of the walls are lower than the center. This ensures that both electrons and holes are confined to the well, provided their energy is low enough [2]. Examples of cavities where this is attainable include GaAs cavities with Al or In-doped GaAs walls [3], and CdTe cavities with Mg-doped CdTe walls [4].

An exciton is an electron-hole pair created by photons exciting the quantum well. Their coupling with the photons introduces an extra term in the Hamiltonian [1],

$$ \mathcal{H}_{\text{exc}} = \int \frac{d^2 k}{(2\pi)^2} \sum_{\sigma} [\bar{\hbar} \omega_{\text{LP}, \sigma}(k) \hat{a}_{\text{LP}, \sigma}^\dagger(k) \hat{a}_{\text{LP}, \sigma}(k) + \bar{\hbar} \omega_{\text{UP}, \sigma}(k) \hat{a}_{\text{UP}, \sigma}^\dagger(k) \hat{a}_{\text{UP}, \sigma}(k)] $$

(6)

Where $X$ and $P$ subscripts now denote excitons and photons respectively, and $\Omega_R$ is the Rabi frequency. Finally, there is a term for exciton-exciton scattering (which is approximated as a contact interaction between field operators $\hat{\psi}_{X, \sigma}(r)$ [1]):

$$ \mathcal{H}_{\text{int}} = \frac{1}{2} \int d^2 r \sum_{\sigma, \sigma'} V_{\sigma, \sigma'}^{XX} \hat{\psi}_{X, \sigma}(r) \hat{\psi}_{X, \sigma}^\dagger(r) \hat{\psi}_{X, \sigma'}(r) \hat{\psi}_{X, \sigma'}^\dagger(r) $$

(7)

### 2.2 The Polariton Hamiltonian

Polariton quasiparticles are a natural consequence of diagonalizing the photon and exciton Hamiltonians. We may write

$$ \mathcal{H}_{\text{cav}} + \mathcal{H}_{\text{exc}} = \int \frac{d^2 k}{(2\pi)^2} \sum_{\sigma} [\bar{\hbar} \omega_{\text{LP}, \sigma}(k) \hat{a}_{\text{LP}, \sigma}^\dagger(k) \hat{a}_{\text{LP}, \sigma}(k) + \bar{\hbar} \omega_{\text{UP}, \sigma}(k) \hat{a}_{\text{UP}, \sigma}^\dagger(k) \hat{a}_{\text{UP}, \sigma}(k)] $$

(8)

Where $\hat{a}_{\text{LP}, \sigma}$ and $\hat{a}_{\text{UP}, \sigma}$ are some linear combinations of $\hat{a}_{P, \sigma}$ and $\hat{a}_{X, \sigma}$ [1]. These are the lower polaritons and upper polaritons respectively. Their dispersion relations

$$ \omega_{\text{UP}, \sigma}(k) = \frac{\omega_{\text{cav}, \sigma}(k) + \omega_{\text{exc}, \sigma}(k)}{2} + \sqrt{\frac{\Omega_R^2}{4} + \left( \frac{\omega_{\text{cav}, \sigma}(k) - \omega_{\text{exc}, \sigma}(k)}{2} \right)^2} $$

(9)

$$ \omega_{\text{LP}, \sigma}(k) = \frac{\omega_{\text{cav}, \sigma}(k) + \omega_{\text{exc}, \sigma}(k)}{2} - \sqrt{\frac{\Omega_R^2}{4} + \left( \frac{\omega_{\text{cav}, \sigma}(k) - \omega_{\text{exc}, \sigma}(k)}{2} \right)^2} $$

(10)
Figure 1: The upper and lower polariton branches in a typical experiment (Image from [2]). At large momenta, lower polaritons resemble pure excitons while upper polaritons resemble pure photons. In an incoherent pumping scheme, the laser first creates excitons at either high energy (red arrow) or high angle (blue arrow), then lets them cool through phonon interactions into the bottom of the lower polariton branch.

... take a distinctive shape in standard experimental conditions, where $\omega_{\text{cav}}(k) > \omega_{\text{exc}}(k)$ is highly parabolic in $k$ and $\omega_{\text{exc}}(k) >> \Omega_R$ is relatively constant in $k$. The upper polariton energy retains the photon’s parabolic band shape while the lower polariton band has the shape of the original exciton band, but with a depression near $k = 0$ (Fig.1). For larger $k$, the upper polaritons resemble pure photons while the lower polaritons resemble pure excitons.

Most experiments involving exciton-polaritons operate in a regime where only the lower polaritons are significant. Near the bottom of the lower polariton band, the dispersion relation gives the polaritons an effective mass on the order of the original photon’s effective mass $m_{\text{cav}}$ - which varies according to Equation 4, but is usually about $10^{-4}$ times the mass of the electron [2],[4].

3 Polariton Condensation

Conventional bosonic gases condense when their thermal de Broglie wavelength

$$\lambda_D = \sqrt{\frac{2\pi\hbar^2}{mk_B T}} \quad (11)$$

stretches beyond the interparticle spacing, specifically when [2]

$$n\lambda_D^3 > 2.62 \quad (12)$$

Crucially, the thermal de Broglie wavelength is proportional to $(mT)^{-1/2}$. Therefore, a lower mass can permit Bose-Einstein condensation at higher temperatures. Due to the incredibly small mass of polaritons, this permits them to condense at much higher temperatures than other particles, even up to room temperature [2].
The data comes from luminescence generated by recombination of polaritons with momentum \( k \), which emits light with wavevector \( k' \) at angle \( \theta \) such that \( k = k' \sin \theta \). Here the threshold power was shown to be around \( 10P_0 \) - at this point, the occupancy of the ground state increased dramatically, as can be seen in the rightmost curve.

In order to demonstrate spontaneous condensation, however, a special type of pumping scheme must be used. As polaritons are ultimately created by laser light, the coherence of the original laser photons needs to be washed out to show that the polaritons spontaneously return to a coherent state. This must be accomplished ([2], Fig.1) in an incoherent pumping scheme, either by creating the excitons at a large angle (and therefore a large \( k \) vector), or by creating them off-resonantly (therefore at a higher energy than normal exciton bands). Either way, the exciton should scatter repeatedly via polariton-polariton or phonon-polariton interactions before they reach the bottom of the lower polariton branch.

Another caveat comes from the dimensionality of the fluid. In the thermodynamic limit, Bose-Einstein condensation in two-dimensional systems can only occur at zero temperature. Quasi long-range order can potentially be created via the BKT transition [2], however it is very difficult to observe characteristics of a BKT transition characteristics, particularly thermally generated vortices. This is partly due to the spontaneous generation of other vortices due to non-equilibrium instabilities [6]. Therefore, all these polariton systems are decidedly finite in size.

### 3.1 Non-Equilibrium Effects

One of the most significant differences between a polariton condensate and an atomic Bose-Einstein condensate, aside from the difference in mass, is that the polaritons occupy a non-equilibrium system (also known as a driven-dissipative system). Polaritons naturally decay with a lifetime on the order of a few picoseconds [2], and are continuously being replenished by the laser. Really, the definitions of a condensate must necessarily be stretched to describe systems like these [7].

The clearest way to represent these non-equilibrium effects is to move to the mean-field picture. Here, the effects become extra terms in the Gross-Pitaevskii Equation

\[
i\hbar \delta_t \psi = \left[ -\frac{\hbar^2}{2m} \nabla^2 + V(r) + U|\psi|^2 \right] \psi
\]  

(13)
We can add gain and loss terms $\gamma$ and $\kappa$ respectively to this equation to obtain

$$i\hbar \delta_t \psi = \left[ -\frac{\hbar^2}{2m} \nabla^2 + V(r) + U|\psi|^2 + i\gamma_{\text{eff}} \right] \psi$$  \hfill (14)

for $\gamma_{\text{eff}} \equiv \gamma - \kappa$, and $m$ is the effective mass at the bottom of the lower polariton band. This would create the problem that condensates will either indefinitely grow or indefinitely shrink depending on the sign of $\gamma_{\text{eff}}$. A more sophisticated way to represent the gain and loss is to couple it with the normal fluid reservoir density $n_R$ [8]:

$$i\hbar \delta_t \psi = \left[ -\frac{\hbar^2}{2m} \nabla^2 + U|\psi|^2 + i\left( R(n_R) - \gamma_{\text{eff}} \right) + 2\tilde{g} n_R \right] \psi$$  \hfill (15)

This equation acknowledges normal fluid polaritons settling into the condensate with the gain $R(n_R)$, and normal-superfluid interactions with $\tilde{g}$. The $n_R$-dependent gain is proportional to the order parameter because it comes from stimulated emission induced by condensed polaritons [8]. The reservoir density $n_R$ is described by the supplementary rate equation

$$\delta_t n_R = P(r) - \gamma_R n_R - R(n_R)|\psi|^2$$  \hfill (16)

This equation includes the natural decay of polaritons in $\gamma_R$, the spatially dependent fixed creation of polaritons (or pump power) $P(r)$, and the conversion of reservoir polaritons into the condensate.

While the exact physics depends on the spatial variation of $P(r)$, generally speaking, there will be some threshold pump power $P_{th}$ below which the $\psi = 0$ solution will be stable, and above which a nonzero order parameter will at least locally form. This order parameter will have an oscillation frequency $\omega$ and a local wavevector $k(r)$, which follow different dispersion relations depending on the pump geometry. Fig. 2 shows an example of the population distribution of the lower polariton branch as $P$ crosses this threshold.

This threshold power has been observed under an incoherent pumping scheme in experiments like [4]. In this experiment, as the pump power $P(r)$ was increased, the occupancy of the $k = 0$ ground state suddenly peaked above the Maxwell-Boltzmann distribution, while the excited state population became saturated. Despite the short lifetime of the polaritons, they found that at high densities, the scattering rate enabled the polaritons to thermalize before they decayed, thus allowing some buildup of the ground state.

### 4 Polariton Superfluid Dynamics

The non-equilibrium Gross-Pitaevskii Equation can be tested by investigating its effects on the dynamics of the polariton fluid. A flowing polariton fluid can be created by a number of coherent pumping schemes, different from the incoherent schemes used to verify polariton condensation. In a coherent scheme, polaritons are driven by a laser and kept at that laser’s energy and momenta without thermalizing,
thus retaining the original coherence of the laser (although some experimentalists
also use an intermediate scheme known as the optical parametric oscillator, where
the polaritons start at the pump momentum and scatter once to a specific signal
point on the lower polariton band [1]). While an incoherent scheme is technically
the more proper approach to creating spontaneously condensing polaritons, the coherent
scheme allows greater control of the fluid’s initial momenta, energy and velocity. In
particular, if the pumping laser hits the cavity at an angle of incidence $\theta$, wavevector
$k_i$, the resulting polaritons will have an in-plane wave vector of $k_0 = k_i \sin \theta$, and a flow velocity [3]

$$ v = \frac{k_0 \hbar}{m} = \frac{k_i \hbar}{m} \sin \theta $$

(17)

Under a purely coherent, spatially invariant, time-independent pump, the phase
of the fluid at all points in space is determined completely by the pump, and non-
trivial features cannot form [1],[9]. In order to observe these features, a coherent
pump either needs to vary its strength over time - such as turning off after some
initial pulse - or it needs to vary its input rate over space - such as only creating
polaritons upstream from a defect [9]. When the flowing fluid intercepts this defect,
it may create perturbations

$$ \begin{pmatrix} \delta \psi(r, t) \\ \delta \psi^*(r, t) \end{pmatrix} $$

(18)

By adding these perturbations to the equilibrium condensate in the non-equilibrium
Gross-Pitaevskii Equation, the following linear equation can be obtained [1]:

$$ \begin{pmatrix} \delta \psi \\ \delta \psi^* \end{pmatrix} = -\mathcal{L}_v^{-1} \cdot V(r) \begin{pmatrix} \phi_0 \\ \phi_0^* \end{pmatrix} $$

(19)

where $\mathcal{L}_v$ is the Bogoliubov operator

$$ \mathcal{L}_v = \begin{pmatrix} -\frac{\hbar^2}{2m} \nabla^2 + U_n & U_n e^{2i\vec{k}_0 \cdot \vec{r}} \\ -U_n e^{-2i\vec{k}_0 \cdot \vec{r}} & \frac{\hbar^2}{2m} \nabla^2 - U_n \end{pmatrix} $$

(20)

And where $\phi_0$ is the unperturbed condensate order parameter, $n = |\phi_0|^2$ is the condensate density, and $V(r)$ is the defect potential [10]. The Bogoliubov operator’s eigenvalues are given by the dispersion relation

$$ \hbar \omega_B(\vec{k}) = \hbar \vec{v} \cdot (\vec{k} - \vec{k}_0) \pm \sqrt{\frac{\hbar^2 (\vec{k} - \vec{k}_0)^2}{2m} \left( \frac{\hbar^2 (\vec{k} - \vec{k}_0)^2}{2m} + 2U_n \right)} $$

(21)

Scattering is possible only if there exists a mode $\vec{k} \neq \vec{k}_0$ such that $\omega_B(\vec{k}) = 0$. The conventional Landau criterion [1] states that this is not possible for $v < v_c$, where the critical velocity $v_c$ is given by
The blue curve at the top is the superfluid regime, where there are no other waves besides \( k_0 \) to scatter too and scattering becomes suppressed. The red curve shows the Cerenkov regime, where the possible scattering states form a ring in momentum space. At the green curve, \( v \approx v_c \), and the energy gradient near \( k_0 \) approaches 0. This means there is a contiguous section of k-space near \( k_0 \) in which the energy difference is small enough to permit scattering. In the case of the experiment in [11], the superfluid density decays over time, which also lowers \( c_s \), hence changing the regime in the direction of the time arrow.

\[ v_c = \min_{\vec{k}} \frac{\omega_B(\vec{k})}{|\vec{k}|} \]  

In the presence of a defect, the critical velocity can be related to the speed of sound \( c_s \) in the fluid. For a local section of the superfluid, \( v_c/c_s \approx 1 \) for small defects, although this does not necessarily mean a subsonic superfluid will always move without dissipation. Large defects can raise the velocity locally, lowering the ratio between the asymptotic critical velocity far from the defect to about 0.37\( c_s \) [1]. In either case, the local Mach number \( v/c_s \) determines the regime the fluid is acting in.

Below the critical velocity, the only possible wavevector \( \vec{k} \) with \( \omega_B(\vec{k}) = 0 \) is \( \vec{k}_0 \) itself, and the fluid does not scatter into other states. This suppression of scattering has been observed experimentally [2], although the superfluid nature of the condensate may not be the best explanation for it [7]. Above this velocity, the possible scattering states form a Rayleigh ring [11] in reciprocal space. The fluid either scatters to states on the edge of the ring, or in the case of \( v \approx v_c \) (the intermediate regime), to states inside the ring itself. In the supersonic regime \( v > c_s \geq v_c \), or the Cerenkov regime, the ring becomes almost circular.
4.1 Observation of Superfluid Dynamics

An important feature of non-equilibrium polariton fluids is that, unless they are specifically prepared in the steady state, their density $\rho$ naturally varies with time. The speed of sound $c_s = \sqrt{\hbar g \rho/m}$ of the fluid increases with the polariton density - this means that even at fixed fluid velocity, an experiment can probe all the different superfluid regimes just by waiting. For example, if the fluid density is allowed to decay without replenishing, there can be a shift from a superfluid to a Cerenkov regime, like in Fig. 3a.

However, there is an even more critical feature of polaritons which makes them a unique environment for studying local superfluid dynamics. The driven-dissipative nature of the condensate, while being problematic in some aspects, makes the local phase and magnitude of the order parameter very easy to observe. When polaritons recombine, they emit light that carries information on the original phase and density of the condensate [3]. By combining this light with a second source of fixed phase on an interferogram, the resulting interference pattern can be used to retrieve this information. For example, a condensate with a uniform phase will produce an interference pattern consisting of rows of parallel fringes [13]. As the velocity of the fluid is related to the gradient of its phase, this even means it is possible to determine the local velocity, and therefore the local Mach number, at any point in space or time [11].

4.2 Vortices and Solitons

Vortices and Solitons are special quasiparticles which can be created when a superfluid exceeds its local critical velocity upon hitting a defect, and they create distinct interference patterns which demonstrate the power of the interferogram observational method. Generally speaking, vortices will be found in the intermediate regime of fluid flow, when there is a large number of possible velocities and $k$ states to form the vortex, while solitons will be found in the Cerenkov regime [11].

A moving superfluid creates vortex-antivortex pairs when there is a tear in its phase front. From [11], this occurs when and where the local Mach number $v/c_s$
exceeds 1, as predicted before. At this point, local fluctuations can temporarily allow phases on either side of the front to connect, creating two topological defects with opposite charge. The vortices can then be identified on an interferogram as fork-shaped dislocations: if the condensate evolves through a phase shift of $2\pi$, an extra fringe will be introduced in the interference pattern, so a mismatch of fringe numbers indicates a phase shift around a vortex core (Fig. 4).

Solitons, specifically dark solitons, are 1-dimensional quasiparticles characterized by a local drop in the superfluid density accompanied by a phase shift on either side. In an interferogram, this can be characterized by a shift in the fringes along the cone behind a defect (Fig. 5). The phase jump $\delta$, soliton density depletion $\rho - \rho_s$, and the soliton velocity $v_s$ are related by the equation [3]

$$\cos\left(\frac{\delta}{2}\right) = \sqrt{1 - \frac{\rho}{\rho_s}} = \frac{v_s}{c_s}$$ (23)

In conventional atomic condensates, solitons are not stable quasiparticles below supersonic speeds. In particular, transverse perturbations often make conventional dark solitons quickly decay into "streets" of vortices along the original soliton valley [14].

In a polariton superfluid, however, the non-equilibrium nature of the system can inhibit this instability [15]. Instead of being ripped apart into vortices, the solitons simply spread out and fade over time, as in Fig. 5. This soliton stability can be seen for speeds as low as $0.6c_s$ [3]. Discrepancies such as this hint at the deeper consequences of the driven-dissipative nature of polariton fluids.

5 Conclusion

Exciton-polariton condensates are a rich avenue of research for studying the dynamics of a quantum fluid, especially in regimes which would normally be difficult to fabricate or observe. These interesting condensates possess a wealth of different phenomena and quasiparticles, of which vortices and solitons are only some examples. Their driven-dissipative nature differs from a conventional condensate and
presents new challenges and curiosities, but also new opportunities to control their creation or measure their local parameters. When used with the proper care and respect for these differences, they can be a powerful tool to understand the physics of superfluids.

References


