

Amplitude-Mode Dynamics of Polariton Condensates

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(Received 11 May 2011; published 20 July 2011)

We study the stability of collective amplitude excitations in nonequilibrium polariton condensates. These excitations correspond to renormalized upper polaritons and to the collective amplitude modes of atomic gases and superconductors. They would be present following a quantum quench or could be created directly by resonant excitation. We show that uniform amplitude excitations are unstable to the production of excitations at finite wave vectors, leading to the formation of density-modulated phases. The physical processes causing the instabilities can be understood by analogy to optical parametric oscillators and the atomic Bose supernova.

DOI: [10.1103/PhysRevLett.107.040401](https://doi.org/10.1103/PhysRevLett.107.040401)

PACS numbers: 05.30.-d, 03.75.Kk, 71.35.Lk, 71.36.+c

In addition to the phase mode responsible for superconductivity and superfluidity, Bose-Einstein condensates possess collective amplitude modes. Examples are the amplitude modes of superconductors [1] and cold atomic gases [2–8], in which the density fluctuates between condensed and noncondensed particles, and the formally equivalent Higgs mode of a relativistic scalar condensate [9]. These modes are orthogonal to the phase modes in the order-parameter space, and their excitation is predicted to lead to nonequilibrium states in which the magnitude of the order parameter oscillates [2–8,10,11].

The strong coupling of excitons and photons in semiconductor microcavities leads to the formation of upper and lower polaritons. Pumping such a microcavity is observed to lead to the formation of Bose-Einstein condensed states of polaritons, characterized by coherent light emission from the structure [12]. This system allows the investigation of physics analogous to that of cold atoms but in a strongly coupled regime with long range interactions mediated by the photons. The coupling of polaritons to electromagnetic radiation outside the microcavity gives an advantage over cold atoms in that polaritons can be coherently controlled by external pumping, and conversely their dynamics and coherence can be directly observed in the emitted light [12,13]. This has allowed experiments to reveal the collective behavior arising from excitations of the phase mode, including superfluidity [14], vortex dynamics [15–17], and the Bogoliubov spectrum [18,19]. However, the collective behavior associated with the amplitude mode has yet to be considered. Controlled excitation of the amplitude mode of an atomic Fermi gas requires a rapid switch of the magnetic field [4–8], while in superconductors a coexisting charge-density wave is needed [1].

The amplitude mode of the polariton condensate may be identified by considering the Dicke model [20,21], which

for weak coupling becomes the BCS model. This limit, with well-known amplitude and phase modes, is adiabatically connected to the strong-coupling limit realized in microcavity experiments, where the collective modes are upper and lower polaritons. Thus the amplitude mode should be identified with the upper polariton. Uniquely, in this system the amplitude mode can be directly driven by resonant excitation, allowing the resulting collective behavior to be studied experimentally. In this Letter, we predict the collective behavior arising when the amplitude degree of freedom of a polariton condensate is manipulated in this way. Whereas driving the phase mode induces superflows, we find that driving the amplitude mode completely destabilizes the condensate, causing the polaritons to spontaneously organize into density-modulated phases (see Fig. 2). This occurs because the interactions transfer the excess rest mass and interaction energy of the nonequilibrium state into kinetic energy, in a way ruled out for the phase modes by the Landau criterion.

We begin by considering the dynamics of a Dicke model of polariton condensation, because for this model exact solutions for the collective dynamics are available [6–8,22]. We show that these solutions, which describe a uniform oscillating condensate, are unstable once excitations at a finite wave vector are considered and systematically identify the instabilities. Our analysis reveals two types (Fig. 1): a wave-mixing instability between the lower and upper polaritons and a modulational instability caused by an attractive interaction between upper polaritons. We propose a Ginzburg-Landau theory which captures these instabilities and provides a realistic model of a microcavity. We use this theory to predict the true steady states under continuous excitation.

We can systematically establish the amplitude-mode dynamics of a polariton condensate by considering the generalized Dicke model [20,21]:

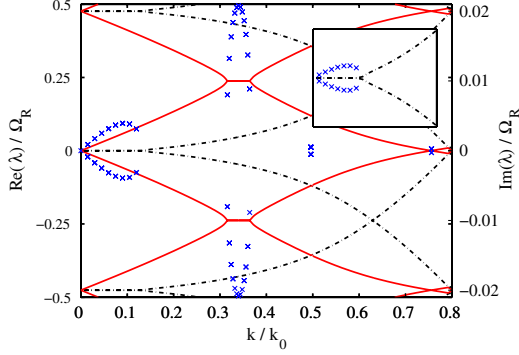


FIG. 1 (color online). Quasienergy spectrum of an oscillating polariton condensate. Lines (left axis) correspond to the real part of the quasienergy λ , and blue crosses to the nonzero imaginary parts (right axis). Modes with nonzero $\text{Im}\lambda$ are unstable. The line colors indicate modes deriving from the phase mode or lower polariton (red solid) or the amplitude mode or upper polariton (black dashed). Inset: Spectrum of a weakly interacting Bose-Einstein condensate with attractive interactions, qualitatively reproducing the small-wave-vector behavior. The parameters are $\omega_0 = -E = 0.4\Omega_R$, $\Delta(0) \approx 0.71$, and $\phi(0) \approx 0.44$, implying a renormalized oscillation frequency $\Omega \approx 0.48\Omega_R$.

$$\hat{H} = \sum_k \omega_k \hat{\psi}_k^\dagger \hat{\psi}_k + \sum_i \frac{E}{2} \hat{\sigma}_i^z + \sum_{i,k} \frac{\Omega_R}{2\sqrt{N}} (\hat{\psi}_k^\dagger \hat{\sigma}_i^- e^{-ik \cdot r_i} + \hat{\sigma}_i^+ \hat{\psi}_k e^{ik \cdot r_i}). \quad (1)$$

This model describes N localized exciton states with positions r_i , in the limit where exciton-exciton interactions exclude double occupancy. Thus each state may be occupied ($\sigma_i^z = 1$) or unoccupied ($\sigma_i^z = -1$). The excitons are coupled to the two-dimensional microcavity photons with in-plane wave vectors k , annihilation operators $\hat{\psi}_k$, and dispersion relation $\omega_k \approx \omega_0 + |k|^2/2m_{\text{ph}}$ ($\hbar = 1$), where $m_{\text{ph}} \approx 10^{-5}m_e$. At this stage we are neglecting important effects such as the dispersion and inhomogeneous broadening of the exciton, the polarization degrees of freedom

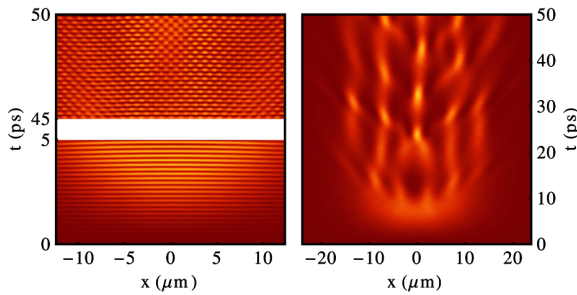


FIG. 2 (color online). Photon density from numerical simulations of Eq. (5), with resonant pumping of both upper and lower polaritons (left panel) and the upper polariton alone (right panel). The left panel shows the formation of a short-wavelength density modulation due to the wave-mixing instability, and the right the result of the attractive-gas instability. The pump field profile is a Gaussian at $x = 0$ with $\sigma = 10 \mu\text{m}$; $\Omega_R = 20 \text{ meV}$; $E - \omega_0 = 4 \text{ meV}$.

[23,24], and the finite lifetime of the cavity photons. We consider only the limit of a single exciton energy E and a single coupling strength parametrized by the Rabi splitting Ω_R . To treat the collective dynamics of (1), it is convenient to work with the expectation values $\psi_k = \langle \hat{\psi}_k \rangle / \sqrt{N}$, $P_k = \langle \hat{P}_k \rangle / \sqrt{N} = \frac{1}{N} \sum_i \langle \hat{\sigma}_i^- \rangle e^{-ik \cdot r_i}$, and $D_k = \langle \hat{D}_k \rangle / \sqrt{N} = \frac{1}{N} \sum_i \langle \hat{\sigma}_i^z \rangle e^{-ik \cdot r_i}$. ψ_k and P_k are the macroscopic components of the electric field and polarization at wave vector k , respectively, while D_k measures the exciton occupation.

With only a single photon mode, the Heisenberg equations for the model (1) are integrable, and closed-form solutions for the dynamics of the collective variables are available [6–8,22]. These solutions have been studied in the context of quench experiments on atomic Fermi gases, in which the pairing interaction would be rapidly switched, giving an impulsive excitation of the collective amplitude mode (analogous experiments are proposed in light-matter systems [10,11]). This is predicted to lead to a spatially uniform condensate in which the order parameter oscillates in time, of which an example is [7]

$$\psi_0 = \phi(t) = \phi_+ \text{dn}(\phi_+ t, \kappa), \quad (2)$$

$$P_0 = \rho(t) = 2[-\omega_0 \phi(t) + i\dot{\phi}(t)]/\Omega_R, \quad (3)$$

where dn is a Jacobi elliptic function and the zero of energy is such that ψ_0 is real. The parameters ϕ_+ and κ determine the period T and magnitude of the oscillations and are known functions of the model parameters and the initial conditions. Note that the oscillation frequency $\Omega = 2\pi/T$ generally differs from Ω_R due to interactions.

Since a microcavity supports a continuum of in-plane modes, we must consider the behavior of an oscillating condensate beyond this mean-field approximation. To do this we linearize the equations of motion about the mean-field solution $\psi_0 = \phi(t)$, $P_0 = \rho(t)$, and $D_0 = \Delta(t)$. The fluctuating parts of the collective variables obey

$$\begin{aligned} i\delta\dot{\psi}_k &= \omega_k \delta\psi_k + \Omega_R \delta P_k / 2, \\ i\delta\dot{P}_k &= E \delta P_k - \Omega_R (\Delta \delta\psi_k + \phi \delta D_k) / 2, \\ i\delta\dot{D}_k &= \Omega_R (\rho^* \delta\psi_k + \phi \delta P_{-k}^* - \phi^* \delta P_k - \rho \delta\psi_{-k}^*), \end{aligned} \quad (4)$$

where the coefficients are time-dependent because the condensate oscillates with angular frequency Ω , which is the gap energy to the occupied amplitude mode. In deriving (4) we assume that the wave vectors are much smaller than the inverse of the spacing of the exciton states, so that motional narrowing is effective and the wave vector of the fluctuations is well defined [25,26].

Equations (4) have the form $\dot{\Psi} = A(t)\Psi$, where Ψ is a vector of the fluctuating fields and $A(t)$ a time-periodic matrix. Thus Floquet's theorem applies, and the solutions are of the form $\Psi = u(t)e^{i\lambda t}$, where $u(t+T) = u(t)$ and the quasienergy λ is defined up to an integer multiple of Ω . Since u is periodic, the stability of the condensate is determined by the imaginary part of λ .

To illustrate the fluctuation spectrum of an oscillating condensate, we show in Fig. 1 the result obtained for the

exact solution (2) and (3). We measure energy and frequency in units of the Rabi splitting Ω_R and wave vector in units of $k_0 = \sqrt{2m_{\text{ph}}\Omega_R}$. The spectrum can be understood by considering the limit of small oscillations around an equilibrium condensate, where (2) becomes $\phi = \phi_0 + \phi_1 \cos \Omega t$ with $\phi_1 \ll \phi_0$, and we have taken the zero of energy to be the equilibrium chemical potential. In this limit the construction of the spectrum is analogous to that of the nearly free electron model [27], with the weak time-periodic component playing the role of the weak periodic potential. To lowest order in the oscillating component, $\phi_1 = 0$, the fluctuation spectrum is that of an equilibrium polariton condensate [20,21]. There is a mode starting at zero energy and wave vector, which in the low-density limit is the lower polariton and in Fig. 1 has been renormalized into the linearly dispersing phase mode. There is also a gapped mode at zero wave vector, which in the low-density limit is the upper polariton [21] and in Fig. 1 is the collective amplitude mode appearing at the gap frequency Ω [28]. The effect of the oscillations is to fold the spectrum in frequency and to couple together the resulting levels. Depending on the phase of the coupling, this can result in either a level repulsion or attraction. In the latter case, the dispersion relation is flattened, and imaginary parts appear for the quasienergies. This signals an instability of the spatially uniform solution and an initially exponential growth of modes at finite wave vectors.

The two strongest instabilities in Fig. 1 occur near $|k| = |k_1| \approx 0.3k_0$ and $|k| \approx 0$. The first occurs where the positive-energy branch derived from the lower polariton crosses with a replica of the corresponding negative-energy branch. In the low-density limit, this occurs only for $E - \omega_0 > 0$. The result is a wave-mixing instability in which an upper and lower polariton from the oscillating condensate at $k = 0$ scatter to a pair of lower polaritons at $\pm k_1$. This is analogous to the instability that drives the microcavity optical parametric oscillator [29], with the two $k = 0$ states forming the pump and the states at $\pm k_1$ the signal and idler. This scattering process has previously been considered as a loss mechanism for incoherent polaritons [30]. The creation of phase modes from amplitude oscillations has been considered in the Bose-Hubbard model [2], and a similar instability has recently been found in the BCS model [31].

The second instability in Fig. 1 corresponds to the flat dispersion relation in the upper polariton branch near $k = 0$. It occurs because the saturation of the light-matter coupling [32] reduces the Rabi splitting with increasing excitation. Thus there is an attractive interaction between upper polaritons, and states containing more than a single such excitation are unstable. The resulting form of unstable spectrum is that obtained from the Bogoliubov analysis for a condensate with weak attractive interactions (inset).

The microscopic theory is capable of describing the initial instability but becomes unwieldy in the nonlinear regime as the unstable modes evolve. To consider the

long-term evolution in a realistic microcavity, we study the Ginzburg-Landau theory

$$\begin{aligned} i \frac{\partial \psi}{\partial t} &= \left(\omega_0 - \frac{\hbar^2}{2m_{\text{ph}}} \nabla^2 \right) \psi + \frac{\Omega_R}{2} (1 - \lambda |P|^2) P \\ &\quad - i\gamma \psi + \xi + F, \\ i \frac{\partial P}{\partial t} &= EP + \frac{\Omega_R}{2} (1 - \lambda |P|^2) \psi. \end{aligned} \quad (5)$$

Here $P(x, t)$ ($\psi[x, t]$) represents the macroscopically occupied exciton (photon) field, which is linearly coupled to the photon (exciton) field to generate polaritons. The nonlinear coupling accounts for the saturation effect. This form can be obtained from (1) by representing the exciton operators with the Holstein-Primakoff transformation and follows directly from the microcavity exciton-photon Hamiltonian [32] treated in mean-field theory. We neglect the Coulomb interactions between the excitons because they act within each quantum well and, hence, are weaker than the saturation nonlinearity when the number of wells is large. This is consistent with the observed redshifting of the upper polariton with density in some microcavities [33]. We have introduced a damping constant $\gamma = 0.25 \text{ ps}^{-1}$ to describe decay of the cavity photons, a term F to describe a resonant pump laser, and a noise source ξ to model spontaneous emission noise. We focus on resonant excitation with circularly polarized light and, hence, include only a single polarization of exciton and photon. For numerical work we set $\lambda = 1$, absorbing the nonlinear coupling strength into the definition of density.

Figure 2 shows the photon densities calculated from (5) in one space dimension, with resonant excitation of both the upper and lower polariton (left panel) and the upper polariton alone (right panel). For these parameters, pumping both modes initially leads to a uniform induced condensate within the pump spot, in which as in (2) the field amplitude oscillates due to intermode beating. However, at later times this uniform state breaks down, and we see the formation of a condensate with a short-wavelength density modulation. We do not see strong signs of thermalization and chaos developing from the instability, as has been suggested for atomic systems [3,31], presumably due to the presence here of dissipation.

In Fig. 2, we also show the behavior when only the upper polariton is pumped. In this case the wave-mixing instability cannot occur, and the dominant instability is due to the attractive interactions. In cold atomic gases, producing a condensate with attractive interactions leads to the Bose supernova [34], where the condensate explodes due to the excess interaction energy of the uniform state. In the microcavity, we instead predict the formation of a large wavelength density modulation. This is because the polaritons can organize in such a way that the excess energy injected by the pump is dissipated, leading to a relatively stable steady state.

While in the undamped model (1) the uniform state is always unstable, the dissipation in (5) implies a threshold density below which uniform states are stable. This density

n_0 is where the gain due to the interactions exceeds the losses from the modes, and therefore for an interaction strength $g \sim \Omega_R \lambda$ is $gn_0 \sim \gamma$, up to numerical factors which are typically of the order of 1. This is essentially the threshold criterion for the optical parametric oscillator [29], so that the threshold density should be within reach experimentally.

The models we have considered can be extended to include more realistic details. In particular, we have neglected the electron-hole continuum, which can produce resonant damping of the upper polariton, similar to the amplitude-mode damping predicted in atomic systems [28,35,36]. However, the two-dimensional exciton binding energy in GaAs (CdTe) is 20 (40) meV, so that the upper polariton in Fig. 2 is below the continuum and not too strongly damped by this mechanism.

An interesting extension of our work would be to allow both polarizations of polaritons and excitons in the Ginzburg-Landau theory. In this case there will be additional amplitude modes connected to fluctuations in the degree of polarization of the condensate. It would be interesting to determine whether the polarization oscillations previously seen in optical parametric oscillator simulations [23] correspond to these amplitude modes and to investigate the possibility of nonlinear decay processes similar to those discussed here. It would also be useful to investigate the possibility of attractive interactions between amplitude modes in the BCS model and, hence, establish the extent to which the instabilities identified here occur in other systems.

In summary, we have used the Dicke model to show that a uniform condensate in which the amplitude mode is excited is unstable due to (a) an attractive interaction between amplitude modes and (b) scattering between amplitude and phase modes. We have used a Ginzburg-Landau theory to show that these instabilities lead to the formation of spatially inhomogeneous condensates. In a microcavity the amplitude mode corresponds to the upper polariton, and therefore these instabilities can be induced by resonant excitation, leading to features in the real-space density and to bright emission at an angle from the cavity. Our work shows that there is a rich collective behavior associated with condensate amplitude modes and that microcavities provide a unique opportunity to explore this physics experimentally.

We acknowledge discussions with J. Keeling and A. O. Silver and support from EPSRC-GB and Science Foundation Ireland (09/SIRG/I1592).

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