Relaxation Oscillations in the Formation of a Polariton Condensate

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We report observation of oscillations in the dynamics of a microcavity polariton condensate formed under pulsed non resonant excitation. While oscillations in a condensate have always been attributed to Josephson mechanisms due to a chemical potential unbalance, here we show that under some localisation conditions of the condensate, they may arise from relaxation oscillations, a pervasive classical dynamics that repeatedly provokes the sudden decay of a reservoir, shutting off relaxation as the reservoir is replenished. Using non-resonant excitation, it is thus possible to obtain condensate injection pulses with a record frequency of 0.1 THz.

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Light-matter particles—that arise from the strong interaction between the photonic field in a semiconductor microcavity and the exciton dipole in quantum wells (QWs)—have recently shown a variety of interesting phenomena related to non-equilibrium condensation. These particles, polaritons, have striking similarities with Bose-Einstein condensates (BECs) of atomic gases, and, in other aspects, with photon lasers. Nonetheless, polaritons have demonstrated peculiarities of their own that set them apart from both quantum condensates and lasers. Recently many observation in fluid dynamics have shown such peculiar behaviors, related to the polaritonic dispersion on the one hand and its dissipative character on the other hand. Fundamental phenomena related to condensation 12, superfluidity 35, quantised vorticity 67, quantum turbulence 89 and phase transitions 10 have shown to deviate from conventional BEC, stimulating new models for the quantum dynamics of polariton condensates. These models consider some of the many parameters that are typical of polaritons, including the upper and lower dispersions, pumping, dissipation, spin, non-linearities, etc. One component that is seldom considered in details is the exciton reservoir. This, however, often plays an important role, even under resonant excitation 1112, and in particular when it comes to polariton relaxation and condensation.

We will show here how a polariton condensate formed by non-resonant excitation, in a confined region of space of a few micron squared, can exhibit marked oscillations of its population. While many oscillatory behaviours have been observed 13 14 or predicted 15 in the polariton literature, relating them to coherent and/or quantum phenomena, we report semi-classical oscillations due to the interplay between reservoir feeding and Bose stimulation. These oscillations fall in the class of so-called “relaxation oscillations” 16, that are well known in class B lasers 17, typically solid-state ones with a small active volume and slow-inversion decay 18, where they lead to the phenomenon of spiking 19. The effect is an important class of self-oscillations 20. In this form, it is analogous to the Tantalus cup oscillations, used by the Romans to measure time, or in natural phenomena such as rhythmic springs. The underlying principle is the following: passed a threshold, a siphon triggers the rapid emptying of a storage that, once depleted, starts over a cycle of refilling under continuous pumping. This versatile physics also takes place in polariton systems in presence of confined states as sketched in Fig. 1 (a). In view of our observations, care should be taken when describing oscillations in small potential wells, when the total population is not conserved but constantly fed from an exciton reservoir. In this case, relaxation and decay cannot be disregarded, being intrinsic to the condensation process.

The experiments have been carried in an Al0.15Ga0.85As/AlAs microcavity with four sets of 3 GaAs quantum wells (QWs) placed at the antinodes of the electric field 21. A pulsed laser of 100 fs excites non-resonantly the polariton population of the lower branch (LPB). A 100X objective lens with a numerical aperture of NA = 0.7 is used to collect micro photoluminescence images resolved in time, space and energy via a streak camera positioned at the end of a 550 mm spectrometer. We use excitation spots of 1.2 µm with a spatial resolution of 0.7 µm in a region of the sample where the amount of inhomogeneities is high enough to
allow for polariton confinement in areas between 1 to 10 µm² and with confining potential of 3 meV in average [22]. Fig. 1 (c-e) shows the dynamics of formation of a condensate at three different excitation powers. When the excitation is below 15 mW, the condensate emission decays with a continuous redshift in time [23] caused by a continuous reduction of the total carriers injected by the laser pulse into the QWs at time \( t_0 \). At powers above \( P_{th} = 15 \) mW, the decay dynamics changes abruptly, showing a striking feature that immediately comes across in Fig. 1(c–e): the formation of the condensate displays an intermittent emission. This oscillatory behaviour can be followed up to three periods at the maximum power, with an almost complete suppression of emission. The time profile of the decays at different excitation powers is shown Fig. 2(a) (black solid line). From Fig. 1 and Fig. 2(a), one can see that the number of maxima is connected to the excitation power and increases for high injection densities. At low pumping powers, the exciton scattering process which feeds the condensate occurs only at early times, giving rise to just one maximum, and resulting in a very short lived condensed state pulse of a few picoseconds. When the number of oscillations is sufficiently high, the long decay tail due to the admixture of exciton recombination and scattering rate is clearly observed. In Fig. 2(c) the oscillation period (full black dots) is plotted together with the initial blueshift (empty red dots), which is proportional to the reservoir population, against the excitation power. There is a monotonous increase of the oscillation frequency when the reservoir population is very high, while for low powers, the oscillations decrease and eventually disappear with low density of the reservoir. Finally, also the rise time depends critically on pumping, as can however be expected for condensation. Unlike cavity photons that are non-interacting, polariton-polariton interactions bring energy into the dynamics of relaxation oscillations, resulting in a redshift in time of the polariton emission.

By choosing a defect of slightly bigger size, the oscillation period shrinks to 10 ps with 4 ps pulse width, as shown in Fig. 3, allowing for the observation of more than ten maxima at a record repetition rate of 0.1 THz. Relaxation oscillations in polariton condensates could be used to drive polariton devices [24] at an extremely fast clock time, which is still longer than the lifetime-limited switching time of polariton non-linearities [25, 26]. Figure 3 also shows several higher energy trails which belong to excited states of the same confined well (see also Fig. S2 in the supporting information). Given that the en-
energy separation is of only 1.2 meV, the condensation of a small fraction of the total population of polaritons is visible for the first, second and third excited state, as was observed in previous works [27–29]. This confirms that the oscillating behaviour is related to the strong confinement effect. While it mainly affects the ground state, in some cases, we observe the presence of oscillations also for the first excited state (Fig. 3).

To show that such oscillations can be explained from the interplay between the exciton reservoir feeding and Bose stimulation only, we describe relaxation through semi-classical Boltzmann equations. The generic equation for the population of the $k$th state reads

$$\dot{n}_k = -\gamma_k n_k + P_k + (n_k + 1) \sum_{k' \neq k} W_{k' \rightarrow k} n_{k'} - n_k \sum_{k' \neq k} W_{k \rightarrow k'} (n_{k'} + 1) \quad [30],$$

with $W_{k \rightarrow k'}$ the scattering rate between states $k$ and $k'$, also with decay rate $\gamma_k$ and pumping rate $P_k$, possibly zero. These equations have been extremely successful to describe the relaxation dynamics of microcavity-polaritons, including condensation [31, 32]. In the case of an infinite system, the quantum number $k$ is the wavevector of a plane wave. In our case, the presence of potentials makes $k$ a label for unspecified eigenstates, which are the solutions of the corresponding Schrödinger equation. To reduce the complexity of the rate equation while retaining the key features of our system, we separate the set of unspecified $k$ states into three groups: the condensate, 0, and two reservoir states, $A$ and $B$, corresponding to the exciton reservoirs in the localized condensation spot ($A$) and outside the condensation area ($B$) [cf. Fig. 1(a)]. Summing over the states $k$ with the assumption that the rates $W_{k \rightarrow k'}$ are identical for $k$, $k'$ in any one of the three groups, and are zero between the condensate and the upper reservoir ($B$), the polariton populations $n_C = \sum_{k \in \{C\}} n_k$ for $C = 0, A$ and $B$ read:

$$\dot{n}_0 = W_{A \rightarrow 0} (n_0 + 1) n_A - W_{0 \rightarrow A} n_0 (n_A + N_A) - \gamma_0 n_0,$$

$$\dot{n}_A = W_{0 \rightarrow A} (n_A + N_A) n_0 + W_{B \rightarrow A} (n_A + N_A) n_B - W_{A \rightarrow 0} n_A (n_0 + 1) - W_{A \rightarrow B} n_A (n_B + N_B) - \gamma_A n_A + P_A \exp(-\gamma_{P_A} t),$$

$$\dot{n}_B = W_{A \rightarrow B} (n_B + N_B) n_A - W_{B \rightarrow A} n_A (n_B + N_A) - \gamma_B n_B + P_B \exp(-\gamma_{P_B} t).$$

Relaxation oscillations also occur with two classes of states only, which is the case of the slow-inversion lasers, where the $\beta$ factor—the ratio of photons that go directly into the lasing mode—determines the oscillatory character [33]. In the polariton case, we find that the equations above can reproduce fairly well the observed dynamics within the main numerical constrain of the problem, that is the one imposed by cavity lifetime, $\gamma_0 = 0.5 \text{ ps}^{-1}$, the only parameter known with high precision. Since our experiment is in the pulsed excitation regime, we have included a decay rate for the pumping terms, $\gamma_{P_A}$ and $\gamma_{P_B}$. In this form, the type of excitation is left in a general form, describing for instance diffusion from remote areas. We have also introduced $N_C = \sum_{k \in \{C\}} 1$, the number of states in each of the group $C$ with $C = A, B$. This simple model accounts for the reservoir and localization as well as drift and diffusion of the excitons. This is enough to reproduce all the qualitative features of the experiment, as shown in Fig. 2(a) (red continuous line), for increasing pumping rate of the higher reservoir $P_B$. One finds that, like in the experimental dataset, the onset of condensation occurs at earlier times with pumping and with an increasing population in the condensate. Passed a given pumping intensity, the condensate buildup and decay is followed by a revival at later times, that also gets closer to the main population peak with increasing pumping and turns into oscillations. At high excitation power, the number of oscillations increases considerably, as can also be seen in the experimental data (Fig. 3). While here we have tuned one parameter only, experimentally, the polariton gas is extremely complex and most parameters do in fact change with pumping [36]. Therefore, our model cannot be expected to achieve more than a qualitative agreement. Nevertheless, allowing more parameters to vary, an excellent quantitative agreement is obtained already at the level of approximation of Eqs. [4], as shown by the dashed lines in Fig. 2(a). The oscillations get washed out mainly due to the increasing rates, both incoming and outgoing, at which the reservoir $A$ is being (de)populated. This effective pumping of the reservoir that directly feeds the condensate can be interpreted as a larger contribution from an increased number of states whose dynamics becomes independent from the oscillations of the condensate. When these oscillations are large, they get imprinted in the reservoir, that
reaches a threshold of stimulated emission leading to its sudden transfer into the ground state until it empties. At this point the condensate is not provisioned anymore and is left to decay, while the reservoir resumes its slower population growth from higher energy states reservoir or provided by diffusion, until it reaches the threshold again, producing another cycle of avalanche into the ground state. Other sensitive parameters are the rates $W_{A\leftrightarrow 0}$ and $W_{A\leftrightarrow B}$ that increase with pumping and account for the main patterns as observed in Fig. 2(b) (to obtain a good agreement, $W_{0\leftrightarrow A}$ and $W_{A\leftrightarrow B}$ are now nonzero).

In what follows, we show how the combination of temporal oscillations and different spatial energy shifts in presence of multiple potential minima can yield not only relaxation oscillations in time but also be combined with a spatial movement. This may give the impression of Josephson oscillations, i.e., spatial oscillations arising from a small population difference between two superfluid regions separated by a potential barrier.

In Fig. 4, we show four snapshots of a polariton condensate which is forming under similar conditions as the ones shown in Figs. 1–3, but where condensation occurs in two areas (separated in space by about one micron) at different times. The whole dynamic of the population during condensation can be better observed in the movie provided in the supplementary material. Figure 4 shows that the condensate first appears in position P1 at early times [Fig. 4(a)], then moves to position P2, which reaches its maximum at time $t = 32$ ps [Fig. 4(b)] and later comes back to position P1 [Fig. 4(c)]. Next it bounces to position P3 [Fig. 4(d)], close to P2, where it reaches again a maximum of intensity at time $t = 90$ ps and finally comes back to P1 where it remains till it decays. Note that for each spatial point, the maxima of emission shift in time according to the condensate position at that time frame. This is due to local difference in the overall redshifts caused by the strong increase and decrease of the polariton population which undergo condensation. This is clear by looking at the emission of points P1, P2 and P3 in Fig. 4(c) as a function of time. While the condensate is forming, the emission maximum shifts in time and so does its position in space, demonstrating a dynamical condensation which is not localised in a single spot. Indeed, together with an oscillating behaviour, which depends on the balance between scattering and depletion of the reservoir, it is also possible to observe a space oscillation which, instead, depends on energy as shown in Fig. 4(e–f), displaying the emission intensity from each spatial position of Fig. 4(a–d) as a function of time (e) and energy (f). In fact, at first the polariton condensate finds most favourable to form in point P1, which has the lowest ground state in the spot area. However, as time increases, the natural redshift—due to a decrease in the total population—becomes less effective where the higher concentration of polaritons is present. This allows the formation of the condensate in more favourable areas (at P2 in this case). Similarly, once the polariton density at P1 has decreased, the condensation tends to form back there, and so on and so forth, giving the impression of synchronized oscillations. This can be evidenced from the redshift of the first and second peaks, at each spatial position, shown in Fig. 4(f). For such inhomogenous condensation area, two simultaneous effects are observed: on the one hand temporal relaxation oscillations, due to the strong localisation of the condensate, and on the other hand, spatial oscillations caused by different blueshift of the condensation regions.

In conclusions, while the relaxation of polaritons has been extensively studied, we demonstrate that, still from the semi-classical standpoint, there remains rich dynamical behaviours to be explored. We have reported for the first time in a polariton system an important type of dynamical instability in a driven dissipative environment, known as relaxation oscillations, present in a large class of systems. This observation is important for future application of polariton as all-optical devices and from a fundamental point of view. They emphasize the crucial role of the exciton reservoir, that can give rise to nontrivial and striking dynamical effects.

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[35] Note that the problem of how to distinguish the polariton regime from its weak-coupling counterpart (lasers) is not proper to relaxation oscillations. This is one of the central problems of the polariton physics, that should be addressed in the same way as when distinguishing, e.g., condensation from lasing: by ensuring that strong-coupling is maintained.

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