Ignition and formation dynamics of a polariton condensate on a semiconductor microcavity pillar

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We present an experimental study on the ignition and decay of a polariton optical parametric oscillator (OPO) in a semiconductor microcavity pillar. The combination of a continuous wave laser pump, under quasi-phase matching conditions, and a non-resonant, 2 ps-long pulse probe allows us to obtain the full dynamics of the system. The arrival of the probe induces a blue-shift in the polariton emission, bringing the OPO process into resonance with the pump, which triggers the OPO-process. We time-resolve the polariton OPO signal emission for more than 1 nanosecond in both real and momentum-space. We fully characterize the emission of the OPO signal with spectral tomography techniques. Our interpretations are backed up by theoretical simulations based on the 2D coupled Gross-Pitaevskii equation for excitons and photons.

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I. INTRODUCTION

Exciton-polaritons in semiconductor microcavities (MCs), when injected with a pump laser close to the inflexion point of the lower polariton branch (LPB) dispersion, undergo a nonlinear process above a pump power threshold. Carrier-carrier interactions self-stimulate a coherent scattering from the pump state into signal and idler polariton states, whose frequency and in-plane momentum fulfill phase-matching conditions. The signal state population, generated by this optical parametric oscillator (OPO)1–4 process, reaches occupation values above one, exhibiting a new form of non-equilibrium superfluid behavior5, metastability of quantum vortices6,7 and persistence of currents8–10.

Lateral etching of planar MCs has been successfully exploited for the creation of a new, large variety of geometries: the resulting discretization of the energy spectrum opens an interesting scenario of different OPO phase-matching conditions in one- (1D) and zero-dimensional (0D) MCs. In the former case, the 1D discretization of the LPB in several energy sub-bands yields the opportunity to obtain exotic intra- and inter-branch OPO processes.11–14 Furthermore, interesting studies of the second order coherence of both signal and idler states have been performed recently.15 In the latter case (0D OPO polaritons), although the most common excitation scheme for micro pillars is non-resonant excitation,16–20 different groups have reported the possibility to induce a parametric oscillation between discrete energy states: from the initial injected energy mode to two neighboring, signal and idler states.21–23

In this work, we study a 40 μm-∅ pillar MC, which is sufficiently large to neglect 0D confinement effects, but still with a bounded spatial extension where polaritons cannot propagate large distances. We excite the sample using a new pump+probe excitation scheme, that differs from common resonant excitation1–4,24. We ignite a long lived OPO polariton signal and observe a transient behavior, characterized by a collective oscillation of the condensed OPO polaritons in the pillar. Thereafter, they reach a quasi-steady state displaying a ring-like emission pattern, due to repulsive interaction with the exciton population in the center of the pillar.25 The OPO signal is switched on with the pulsed probe at the exciton energy level which blueshifts the LPB making the continuous wave (cw) pump enter in resonance conditions in a OPO-process that lives for ∼1 ns. We study the full dynamics of the creation and decay of this confined OPO condensate in real and momentum-space (k-space). The interpretation of the experimental measurements of the spatial emission dynamics is supported by theoretical simulations using the 2D coupled Gross-Pitaevskii equations for excitons and photons.

II. SAMPLE AND EXPERIMENTAL SETUP

We investigate a high-quality 5λ/2 AlGaAs-based MC with 12 embedded quantum wells, and a Rabi splitting...
\( \Omega_e = 9 \) meV. Further information about this sample is given in Ref. 26. Pillars with different diameters have been sculpted through reactive ion etching. We have chosen a 40 \( \mu \text{m} \)-\( \varnothing \) pillar in an area of the sample where the detuning is close to zero.\(^{27} \) Figure 1 shows a scanning electron microscopy image of such a pillar, including the excitation scheme.

The sample, mounted in a cold-finger cryostat and kept at 10 K, is excited with pump and probe laser beams under the following conditions. For the continuous wave experiments, Section III A, we use only a pump beam obtained from a \( \text{cw} \) Ti:Al\(_2\)O\(_3\) laser. It is tuned at \( E_p = 1.5416 \) eV, impinging on the sample with an in-plane momentum \( (k_p)_z = -1.9 \) \( \mu \text{m}^{-1} \), fulfilling the phase-matching conditions \( 2E_p = E_s + E_i \) and \( 2k_p = k_s + k_i \) (where the subindex \( s/i \) means signal/idler). Its power is set to \( P_p = 160 \) mW. For the time-resolved experiments, Subsecs. III B and III C, the excitation scheme is represented in Fig. 2. In this case the \( \text{cw} \) pump beam is out of OPO phase-matching conditions, since now its energy is tuned slightly above the LPB \( (E_p = 1.5419 \text{ eV}) \). The second excitation source, probe, is a pulsed Ti:Al\(_2\)O\(_3\) laser (2 ps-long pulses); it is tuned into resonance with the exciton mode \( (E_{ph} = 1.5445 \text{ eV}) \), and its power, \( P_{ph} = 230 \) mW, is strong enough to trigger the OPO-process together with the pump. The origin of time \( t = 0 \) is set to the instant when the probe impinges on the pillar. The two excitonic lines labelled \( X_1 \) and \( X_2 \) originate from excitons uncoupled to the cavity modes, due to slight quantum well thickness variations, of the order of a monolayer, between different quantum wells. Further information about the origin of the exciton emission is given in Ref. 25.

For the experimental results described in Sec. III, the laser beams are focused on the sample through a high numerical-aperture (0.6) lens, forming two overlapping elliptically shaped spots (10/20 \( \mu \text{m} \)-\( \varnothing \) minor/major axis along the Y/X axis) impinging on the pillar with an in-plane momentum \( k_{p/\text{ph}} = \{ (k_{p/\text{ph}})_x, (k_{p/\text{ph}})_y \} = \{ -1.9, 0 \} \mu \text{m}^{-1} \), see full arrows in Fig. 1. The same lens is used to collect and direct the emission towards a 0.5 m spectrometer coupled to a CCD (Section III A) and a streak camera (Subsecs. III B and III C). The photoluminescence (PL) can be resolved in the near-\( (k\)-space) as well as in the far-field (\( k\)-space). The distribution of polaritons in \( k\)-space is accessed by imaging the Fourier plane of the lens used to collect the PL, taking advantage of the direct relation between the angle of emission and the in-plane momentum of polaritons.\(^{28} \) To avoid the direct reflection of the pump and probe beams, we block the emission in \( k\)-space for \( |k| > 1.5 \mu \text{m}^{-1} \) and we spectrally filter the polariton PL with an energy detection range of 1 meV around 1.54 eV. The lens that focuses the real or \( k\)-space PL distribution into the spectrometer entrance slit is displaced laterally by discrete steps, yielding a tomographic reconstruction of energy- and time-resolved images.

### III. EXPERIMENTAL RESULTS AND DISCUSSION

This section compiles the results of \( \text{cw} \) and time-resolved experiments in the pillar, which are organized as follows. In Section III A we address the tomographic spectral distribution of polaritons in the pillar under
cw-OPO excitation, in both real- and k-space. In Section III B we study the dynamics of the polariton emission under pump+probe excitation. In this case, the cw pump laser beam is out of OPO-conditions (slightly blue-detuned from the LPB); only after the arrival of the probe, the induced blue-shift of the LPB is large enough to start the OPO-process. In Section III C, for the sake of completeness, we present the polariton dynamics with the probe excitation only, where the decay of the polariton PL is faster and limited by the relaxation dynamics from photo-generated excitons towards the polariton ground state.

A. CW spectroscopy characterization of OPO signal emission

Figure 3 summarizes the spectral tomography of the polariton emission in both real- and k-space under a cw pump excitation. The OPO signal energy is $E_s \approx 1.54$ eV, with a full width $\Delta E_s \approx 0.8$ meV. Figure 3(a)/(e) shows the polariton emission in real-/k-space (Y/k_y) at the central X = 0/k_x = 0 cross-section in the pillar. The three, dashed, vertical lines mark the position where the full X = Y/k_x - k_y PL map has been reconstructed in panels (b-d)/(f-h). The three selected energies are: $E_1 = 1.5398$ eV, $E_2 = 1.5400$ eV and $E_3 = 1.5402$ eV. In panels (b-d) dashed, red circles mark the limits of the pillar. A vertical red line marks the resolved cross-section of the spectrum shown in panels (a)/(d) for real-/k-space. The PL maps are coded in linear, false color scales.

We perform a two-dimensional reconstruction of the spectrum emission, showing the full X = Y/k_x - k_y PL distribution of polaritons at three, selected energies: $E_1 = 1.5398$ eV, $E_2 = 1.5400$ eV and $E_3 = 1.5402$ eV, Figs. 3(b-d)/(f-h). Figure 3(b) reveals the ring-like distribution of polaritons at $E_1$. The angle of incidence of the pump creates an asymmetrical blueshift in the left side of the pillar due to polariton propagation, so the polariton ring-shaped emission is broken in the region $\{x, y\} = \{-10,0\} \mu m$. At a higher energy $E_3$, Fig. 3(c), polaritons emit from a ring of radius 10 $\mu m$ with a smaller side gap. Figure 3(d) shows an almost flat disk of emission, whose radius is $\sim 15 \mu m$.

In k-space, Fig. 3(f) shows a small disk (radius 0.7 $\mu m^{-1}$) in the polariton distribution. This demonstrates that polaritons lying at low energy ($E_1$), which are distributed along the ring in Fig. 3(b), are confined in real space, with a small amount of kinetic energy. The confinement of polaritons at $E_1$ is also hinted in the momentum cross-section in Fig. 3(e). Figures 3(g) and 3(h) evidence a flat distribution in momentum space, where polaritons have all possible values of momenta inside a disk of radius |k| < 1 and 1.5 $\mu m^{-1}$, respectively. For energies higher than $E_3$, polariton emission is distributed in a ring (not shown) corresponding to a cloud of uncondensed, hot polaritons in the LPB.

B. Igniting a long-living OPO-process with a probe-induced blueshift

In this section we show how the OPO process is triggered by the arrival of a probe beam. Moreover, through real-space measurements, we reveal the ring-shape distribution of signal polaritons and, analyzing k-space images, we characterize their movement around the pillar.

The pump (cw) + probe (pulsed) excitation configuration activates a long-lived stimulated OPO scattering process. The time during which the OPO is active is much longer than the photon lifetime, estimated from the Q-factor to be $\sim 10$ ps. The real- and k-space dynamics or the signal emission are presented in Figs. 4 and 5, respectively. In these time-resolved experiments, the energy resolution is similar to that of the energy width of the OPO signal $\Delta E_s$.

Figure 4 compiles two dimensional images of the polariton signal emission at different times in real-space. For each panel, the time is displayed at the left upper corner, being the temporal origin set at the instant when the probe impinges on the sample. Fig. 4(a) shows that, before the arrival of the probe pulse ($t = -99$ ps), there is no signal emission since the pump is off of phase-matching conditions. At $t = 0$, Fig. 4(b), the probe impinges on the pillar; the spot shape is distorted due to the fact that $k_{pb} \neq 0$. The polariton emission rapidly arises from the whole pillar surface, seen as a flat homogeneous disk, Fig. 4(c). The PL increases during $\sim 100$ ps, as shown in Fig. 4(d); thereafter, the extra-population of polaritons induced by the probe decreases. When the OPO process has switched-on, the polariton dispersion becomes ring-shaped, Fig. 4(e): the real-space polariton distribution resembles that shown in Fig. 3(c). Polaritons emit close to the border of the pillar, due to the blueshift induced by...
FIG. 4. (Color online) Real-space polariton emission dynamics in the pillar under pump and probe beam excitation. The time is shown by the labels in each panel. The time $t = 0$ coincides with the arrival of the probe. The PL is coded in a linear, false color scale shown on the right of each panel. The complete $X - Y$ polariton emission dynamics is available as Supplemental Material.\textsuperscript{29}

the cw pump. This polariton emission persists for more than $\sim 1$ ns, see Figs. 4(f-h).

Figure 5 shows the dynamics in k-space of the polariton population. As observed in real-space, there is no OPO signal emission before the arrival of the probe, Fig. 5(a). The pulsed probe arrives to the pillar at $t = 0$, Fig. 5(b), the spurious emission from $k \sim 0$ is unfiltered scattered laser light. The probe creates an extra polariton population around $k = \{-1.5,0\} \, \mu$m$^{-1}$ that rapidly moves in the $+k_x$ direction (not shown here, see the movie in the Supplementary Material).\textsuperscript{29} At $t = 34$ ps, Fig. 5(c), the population has moved towards $k = \{1.5,0\} \, \mu$m$^{-1}$. Fig. 5(d) shows that, at $t = 50$ ps, the polariton emission is homogeneously distributed in a disk of radius $|k| \approx 1 \, \mu$m$^{-1}$. At $t = 67$ ps, Fig. 5(e), the population reverts its angle of emission towards $-k_x$. As it was mentioned for Fig. 4, $\sim 100$ ps after the arrival of the probe, its induced extra-population weakens and the OPO signal is redistributed at the center of k-space, Fig. 5(f). The oscillations arise from the probe-injected excitons, which are excited with a certain angle. The relaxation of excitons yields polaritons that possess a non-zero momentum. As clearly inferred from the dynamics of the k-space distribution, polaritons change their initial momentum due to several bounces against the MC wall. The gain follows the injected probe distribution as it moves within the pillar, until the excitonic population dies off and a more stable, switched-on OPO process takes place, which resembles the polariton distribution in real and k-space shown in Fig. 3. For longer times, $t = 212$ ps (Fig. 5(g)), the emission is mainly perpendicular to the sample surface, i.e. at $k \approx 0$. At later times, Fig. 5(h), two effects are observed: firstly, there is a lobe-like structure at $k = \{1.0,0\} \, \mu$m$^{-1}$ and, secondly, there is a progressive decay of the central ($k = 0$) population. The latter effect is due to the fact that the emission energy is red-shifting with time and we have not been following this red-shift with the streak camera since its energy detection is fixed for these experiments.

C. Non-resonant excitation solely with a pulsed probe

In this section, for completeness, we address the polariton dynamics obtained when only the pulsed probe beam excites the pillar. The polariton emission dynamics in real- and k-space is summarized in Figs. 6(a-d) and Figs. 6(e-h), respectively. Fig. 6(a) shows the absence of emission before the arrival of the probe. Figs. 6(b,c) show the switch-on of the polariton emission, following a similar dynamics to that shown in Fig. 4, during the first $\sim 100$ ps. Fig. 6(d) demonstrates the shorter lifetime of the polariton population created under these excitation conditions; after $\sim 250$ ps, the polariton PL has disappeared.

In a similar fashion, Figs. 6(e-h) show the dynamics of the emission in k-space. An oscillation of the polariton momentum similar to that described in Fig. 5(c-e) is obtained here during the first $\sim 100$ ps. The polariton emission moves in the $k_x$ direction, going from $-1.5 \, \mu$m$^{-1}$ to $+1.5 \, \mu$m$^{-1}$ in $\sim 70$ ps. At later times, Fig. 6(g), the emission originates from $k \approx 0$, and for $t = 355$ ps, the lack of emission is confirmed, Fig. 6(h).

D. Comparison of the dynamics of the two excitation schemes involving a probe

The dynamics of the spatially integrated emission build up is depicted in Fig. 7. A similar behavior is obtained for pump+probe (blue line) and probe-only (orange line) excitation conditions. The PL reaches its maximum emission $\sim 50$ ps after the probe is gone. It is on the decay dynamics that differences between the two excitation conditions appear. Under probe-only excitation we observe a
FIG. 5. (Color online) Momentum-space polariton emission dynamics in the pillar. Same excitation conditions as in Fig. 4. The time is shown by the labels in each panel. The PL is coded in a linear, false color scale shown on the right of each panel. The complete $k_X - k_Y$ polariton emission dynamics is available as Supplemental Material.\textsuperscript{29}

FIG. 6. (Color online) (a)-(d) Real/(e)-(h) $k$-space polariton emission distribution in the pillar, the time is shown by the labels in each panel. Only the probe beam excites at the center of the pillar. The time $t = 0$ coincides with the arrival of the probe. The PL is coded in a linear, false color scale shown on the right of each panel. The complete $X - Y$ and $k_X - k_Y$ polariton emission dynamics is available as Supplemental Material.\textsuperscript{29}

mono-exponential decay of the PL, with a characteristic decay time of 63 ps. The decay is markedly different in the presence of the out-of-resonance pump. In this case, the PL decay is bi-exponential, with a fast decay time of 76 ps, and a long-lived polariton population lasting more than 1 ns, evidenced by the slow, $\sim 500$ ps, decay time.

IV. THEORETICAL DESCRIPTION

To model our experimental results under the OPO configuration, described in Section III B, we make use of the 2D coupled Gross-Pitaevskii equations for photons $\psi(x, y)$, Eq. 1, and excitons $\varphi(x, y)$, Eq. 2:

$$
i \hbar \frac{\partial \psi}{\partial t} = -\frac{\hbar^2}{2m_{ph}} \Delta \psi + \frac{\hbar \Omega_R}{2} \varphi + \frac{\hbar}{2\tau_{ph}} \psi^* \psi + P + P_X + f$$  \hspace{1cm} (1)$$

$$
i \hbar \frac{\partial \varphi}{\partial t} = -\frac{\hbar^2}{2m_X} \Delta \varphi + \frac{\hbar \Omega_R}{2} \psi + \alpha_1 |\varphi|^2 \varphi + U \varphi$$ \hspace{1cm} (2)$$

Here, $m_{ph} = 4 \times 10^{-5} m_0$ is the photon mass, $m_X = 0.6 m_0$ is the exciton mass ($m_0$ is the free electron mass),
when the probe excitons have decayed through relaxation non-resonant spontaneous scattering creates 0.01 particles. To describe the cell of pumping provides an average of 10 particles in a unit cw to account for the effects of spontaneous scattering. The f to the polariton branch, and values as in the experiments), blue-detuned with respect the tem at a given frequency and in-plane momentum (same values as in the experiments), blue-detuned with respect to the polariton branch, and f is the noise, which serves to account for the effects of spontaneous scattering. The cw pumping provides an average of 10 particles in a unit cell of h = 0.25 µm in the steady state, while the spontaneous scattering creates 0.01 particles. To describe the non-resonant probe, we use a pulsed pumping term PX, tuned at the exciton resonance, with the same duration and wavevector as in the experiments. No disorder potential was taken into account, because its effects were not observed in the experiments.

The confinement potential of the pillar, acting on the photon and excitonic parts, is described by U. τph = 1 ps is the photon lifetime (the exciton decay is neglected), P is the quasi-resonant pumping term, exciting the system at a given frequency and in-plane momentum (same values as in the experiments), blue-detuned with respect to the polariton branch, and f is the noise, which serves to account for the effects of spontaneous scattering. The cw pumping provides an average of 10 particles in a unit cell of h = 0.25 µm in the steady state, while the spontaneous scattering creates 0.01 particles. To describe the non-resonant probe, we use a pulsed pumping term PX, tuned at the exciton resonance, with the same duration and wavevector as in the experiments. No disorder potential was taken into account, because its effects were not observed in the experiments.

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favorable for the OPO only in these points, located at the minima between the pump spot and the pillar boundary: the pump density at the center of the pillar is too high to maintain resonant OPO, because the signal polaritons are pushed away from the center and there their density becomes insufficient to maintain stable OPO. The overall good agreement between theory and experiment supports our interpretation of the experimental observations.

V. CONCLUSIONS

We have presented new experimental conditions to obtain a long-lived polariton condensate in a pillar MC. It involves two excitation beams impinging at the center of the pillar with the same wavevector: a cw pump, slightly blue-detuned from the inflection point of the LPB, and a pulsed probe, resonantly creating excitons. The polariton population created with the arrival of the probe induces a blue-shift of the LPB, which enters into resonance with the pump beam, triggering the OPO-process. The cw pump keeps on feeding the OPO after the pulsed probe has disappeared, because of the hysteresis of the polariton bistability. As a result of the combined effect of both beams, the OPO signal emission lasts for more than 1 ns, much longer than any of the characteristic times of the MC. The polariton condensate dynamics observed when using just the probe beam is remarkably similar, but much shorter lived, to that obtained for the two beam excitation. The exciton population created by the probe beam efficiently relaxes to the LPB and from there follows the OPO dynamics. The characteristic decay time is one order of magnitude shorter than that obtained under the two beam excitation conditions.

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We define the exciton–photon detuning as $\delta = E_C - E_X$, where $E_C$ and $E_X$ are the cavity mode and exciton energy at $k = 0$, respectively.