

Synchronized and Desynchronized Phases of Exciton-Polariton Condensates in the Presence of Disorder

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Condensation of exciton polaritons in semiconductor microcavities takes place despite in-plane disorder. Below the critical density, the inhomogeneity of the disorder limits the spatial extension of the ground state. Above the critical density, in the presence of weak disorder, this limitation is spontaneously overcome by the nonlinear interaction, resulting in an extended synchronized phase. In the case of strong disorder, several non-phase-locked condensates can be evidenced. The transition from a synchronized phase to a desynchronized phase is addressed by sampling the cavity disorder.

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Quantum phase transitions realized in inhomogeneous potentials is a topic of major importance in condensed matter physics. It involves nontrivial matter states, such as Josephson oscillations for double potential wells [1], the Mott insulator for periodic potentials [2], and Bose glass for disorder potentials [3]. The case of disorder potentials is a crucial matter both for the intrinsic interest of phase transitions and for the unavoidable disorder in real systems [4]. The recent achievement of Bose-Einstein condensation for microcavity polaritons [5] addresses again this issue. Polariton condensation indeed takes place in a disordered medium that creates traps, in which the polaritons are preferentially located.

Polaritons are two-dimensional (2D) quasiparticles, resulting from the strong coupling of cavity photons with quantum well excitons. Their very low mass allows the observation of condensation at high temperatures. Because of their short radiative recombination time, the phase transition gets a nonequilibrium character [6]. The fundamental difference with equilibrium phase transitions is that here the stationary state corresponds to a balanced flux of incoming and outgoing particles in the condensate. The cw pump laser intensity plays the role of the driving parameter of the phase transition. Because of their excitonic part, polaritons can interact. At the densities necessary to achieve condensation, these interactions happen in the dilute limit [7] and are evidenced through the spectral blueshift of the states. To these two intrinsic features of such light-matter quasiparticles is added a particularity of the II-VI semiconductor microcavity that displays cavity length fluctuations [8]. This results in strong fluctuations of the polariton energies over small distances, which create some sort of trap for polaritons. Eventually, the nature of the condensed phase will be the result of the complex interplay between disorder effects, linear coupling, and nonlinear interactions within the very short lifetime of the polaritons.

In this Letter, we focus on the role of disorder on the spontaneous formation of a 2D extended polariton conden-

sate. This issue differs from the one discussed in atomic systems [9], where the condensate is created in the absence of disorder and eventually experiences a disordered potential. At low densities, the initial polariton cloud is already subject to the in-plane disorder which is responsible for spectral fluctuations of the polariton states. We shall give experimental evidence that, at high polariton density, nonlinear interactions—manifested by the polariton blue-shift—are necessary for the formation of a synchronized phase, i.e., the macroscopic occupancy of a single coherent quantum state—delocalized over several potential traps—not originally existing. This site to site synchronization is demonstrated by coupled spectral and interferometric measurements. To our knowledge, this is the first experimental evidence of such a frequency locking for a polaritonic system undergoing a phase transition. For some configurations of the disorder depending on the position on the sample, a desynchronized phase with several nonsynchronized condensates is reached, corresponding to a nonuniform order parameter.

Synchronization is a general behavior observed in various physical systems since the historical Huygens coupled pendulums [10]: arrays of Josephson oscillators [11], arrays of vertical-cavity surface-emitting lasers [12], or the microwave spin torque nano-oscillators [13]. The basic idea of this effect is that initially independent oscillators end up sharing a correlated evolution; i.e., they are linked by some phase relation, whether their frequencies are equal or not. This can be achieved by an internal or an external force, resulting in scattering or injecting some portion of one oscillator into its neighbors. The oscillators participating in a synchronized system must possess a nonlinear response to force synchronization.

Our study of the condensate formation reveals two different cases, one for which a synchronized phase is reached despite disorder and one for which nonlinear interactions do not allow the existing disorder to be overcome. These two different behaviors will be highlighted by a set of experiments implemented at two different positions

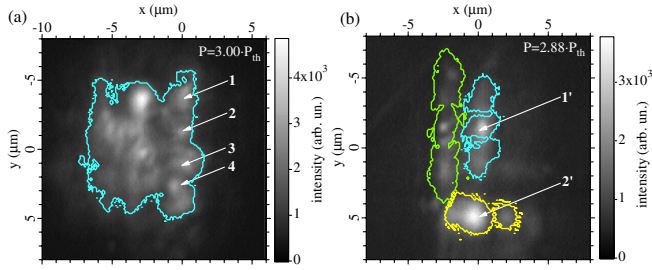


FIG. 1 (color online). (a) Real space luminescence of one single condensate taken at position 1 and (b) three independent condensates taken at position 2.

1 and 2 (Fig. 1) that differ only by the local potential disorder.

The sample is the same one as in our previous works [5,8]. The excitation is performed in a nonresonant quasi-cw way. Spatial and spectral resolutions are achieved by using a real space imaging setup in conjunction with a $10 \mu\text{eV}$ resolution spectrometer. The diffraction limited real space image of the sample surface is formed on the entrance slit of the spectrometer. This allows us to select one line ($x = 0$) in real space and to resolve spectrally the luminescence from each point of this line (Fig. 2). Such an image contains no information on the angular emission from the microcavity since the emitted light is integrated from -30° to 30° . Let us stress that the ground state (that emits light at normal incidence) and all of the excited states contribute to the measured luminescence.

The integrated luminescence emitted by any area of the sample appears to be homogeneous for low pump intensities [5,8]. This changes dramatically when we spectrally resolve the signal. For position 1 [Fig. 2(a)], we observe clear fluctuations of the ground state energy, which are due to the disorder of the sample structure. At low densities, we clearly observe a nonthermal distribution with a bottleneck effect on the lower polariton branch dispersion (not shown). When the pump power is increased towards the condensation threshold, the bottleneck disappears, and the polariton cloud becomes thermalized at 19.2 K due to increased polariton-polariton scattering. As the pump is increased above threshold, two pronounced effects are evidenced at position 1: (i) There is a common blueshift across the whole excitation region; (ii) a spectrally homogeneous delocalized ground state appears over the entire excitation region.

To demonstrate unambiguously that this single ground state is defined only above threshold, we put in evidence the first-order correlations by means of a modified Michelson interferometer [5] with spectral resolution along the line studied in the previous experiment. The region of significant contrast away from the central peak of autocorrelation [Figs. 2(c) and 2(d)] shows the spatial extension of the ground state. At higher energy $E(|\mathbf{k}|)$, all of the states of the same wave vector \mathbf{k} in modulus contribute, and correlations vanish. Below threshold

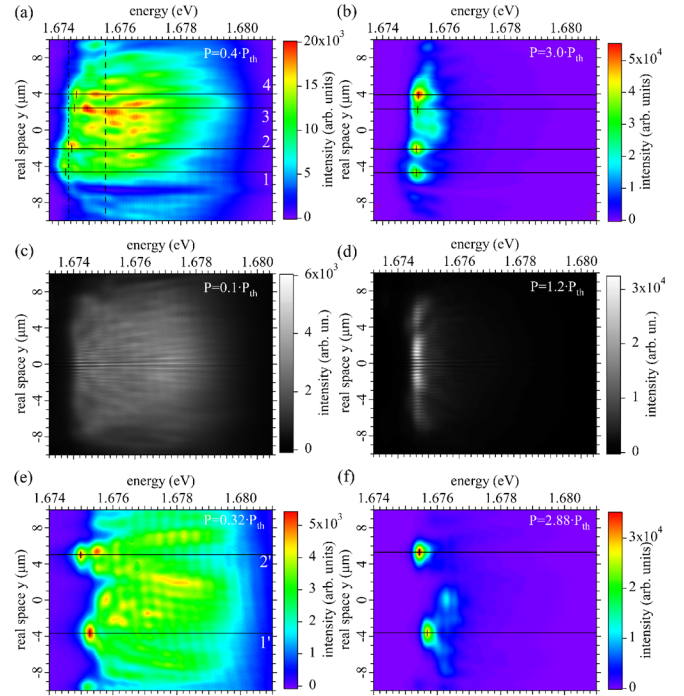


FIG. 2 (color online). (a)–(b) and (e)–(f) Spectrally resolved real space line and (c)–(d) interferogram. Horizontal axis: Energy in electron volts. Vertical axis: Real space in microns. (a) Profile along $x = 0$ line in Fig. 1(a) (position 1), at $P = 0.4P_{\text{thr}}$. At the same position, the ground state energy given by the spatially integrated dispersion curve is at 1.67433 eV (dashed-dotted vertical line). The bottleneck appears on the same dispersion curve around 1.67554 eV (dashed line). Small vertical lines: Energy of the ground state. (b) The same at $P = 3P_{\text{thr}}$. (c) Spectrally resolved interferogram at $P = 0.1P_{\text{thr}}$ and (d) the same at $P = 1.2P_{\text{thr}}$. (e) Profile along $x = 0$ line in Fig. 1(b) (position 2), at $P = 0.32P_{\text{thr}}$. (f) The same at $P = 2.88P_{\text{thr}}$.

[Fig. 2(c)], the spatial extension of the first-order coherence is limited by disorder to a few microns: There is no common ground state for polaritons localized in different potential minima. This experiment rules out any description exclusively formulated in terms of linear coupling, which should define one single ground state irrespective of the excitation density. Above threshold, the coherent domain reaches the size of the area excited by the laser in the case of position 1 (this enhancement is reduced in the case of position 2; see below).

We investigate the evolution of the frequency detuning of spatially separated luminescent spots of the condensate [Fig. 1(a)] across the condensation transition. The energy of the ground state is obtained by using a Lorentzian fit on the low energy side, with a fixed linewidth corresponding to the ground state ($350 \mu\text{eV}$, measured separately). The frequency detuning is then defined as the frequency difference between the energies of the fitted Lorentzians at the two different positions. We selected four luminescent points [see Fig. 1(a)] and monitored the frequency detuning for the different pairs of neighbors (see Fig. 3). When

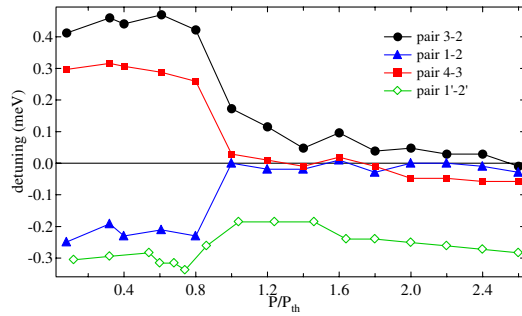


FIG. 3 (color online). Frequency detuning for different pairs of spatially separated polaritons as a function of the excitation intensity. The experimental points are connected by lines. Points 1, 2, 3, and 4 belong to a single condensate (position 1), and points 1' and 2' belong to independent condensates (position 2).

increasing the density over the critical value, we observe a very clear reduction of the frequency detuning for all pairs of neighbors: The initial detunings are of the order of $300 \mu\text{eV}$ below threshold and become smaller than $75 \mu\text{eV}$ above threshold. This residual detuning is very small compared to the linewidth of the condensate (at minimum $400 \mu\text{eV}$). It is also negligible when compared to the global blueshift, which is $500 \mu\text{eV}$ between P_{th} and $3P_{\text{th}}$.

In order to relate this transition with the formation of a polariton condensate, we have to consider again the spatial coherence properties. We concentrate here on a 2D mapping of $g^{(1)}(\mathbf{r}, -\mathbf{r})$ [5]. The region over which correlations are significant defines the condensate domain. For position 1, it demonstrates that all of the points for which we observe a clear reduction of the frequency detuning belong to the same condensate [Fig. 1(a)].

From the experiments presented above, we conclude that the formation of extended 2D polariton condensates involves a nonlinear interaction. It can come both from condensate-condensate interactions and from the reservoir of excitons and polaritons in the condensate [14]. Nonetheless, this does not exclude a linear coupling such as, for example, photonic coupling through the potential barrier, necessary for the synchronization to occur. Our understanding is that nonlinear interactions allow, via the blueshift of the states, the reduction of the initial detuning between polariton states corresponding to different potential minima, up to the point where the linear coupling can operate to induce synchronization. Experimentally, a study of the contribution of these different effects would require full control of the disorder potential. Here we can give only the characteristics of this potential: The relative distance between neighbor potential minima varies from 2 to $6 \mu\text{m}$, the relative difference of the potential depths shows large fluctuations up to $500 \mu\text{eV}$, and the shape of the traps is irregular. These features explain why the possibility to get a synchronized phase depends strongly on the position on the sample [15].

We have repeated the same experiments on position 2, at the same exciton-photon detuning (between 0 and 3 meV), and the general behavior differs dramatically. The diamond points (Fig. 3) present the evolution of the frequency detuning between polaritons at two different positions 1' and 2' [Fig. 1(b)]. They start with an initial frequency detuning of $300 \mu\text{eV}$, which decreases slightly at the critical density but remains significant at high densities. Thus, although this frequency detuning is lower than the linewidth and the blueshift, there is no synchronization, as demonstrated in Fig. 2(b). This suggests that condensed polaritons at 1' and 2' are independent. This can be evidenced through a proper mapping of the correlations, as described above. It is well known that, similarly to the case of two independent lasers [16], two independent condensates will interfere during the shortest coherence time [17]. Considering the 6 ps coherence time of the polariton condensate [5], the interference between independent polariton condensates will never be observable. Thus, the lack of interference between condensed polaritons in two different potential wells can be used as a direct proof that they belong to two independent condensates (let us note that in this case a spectrally resolved interferometric measurement is not relevant). By repeating the mapping experiment at proper positions, we were able to identify three independent condensates at position 2. As expected, polaritons at positions 1' and 2' belong to two independent condensates. Let us stress that they have a non-negligible spectral overlap; thus, it is meaningful to probe if they are correlated or not, via an interferometric measurement.

We finally end up with two opposite cases for the condensation of polaritons localized in neighboring potential wells: Either they merge into one single condensate or they separate into independent condensates. To study the transition between these two cases, we choose to discriminate between dependent and independent condensates by comparing the frequency detuning in the condensed phase $\Delta\nu$ with one parameter: the initial frequency detuning $\Delta\Omega_0$. Pairs of polaritons belonging to one single condensate will appear on the $\Delta\nu = 0$ line and independent condensed polaritons on the $\Delta\nu = \Delta\Omega_0$ line. This approach is a standard practice in the description of systems of coupled oscillators, introduced initially by Winfree [18] and Kuramoto [19] in biological systems exhibiting mode synchronization. It has been theoretically extended and applied, for example, to fireflies (collective flashing) and crickets (chirp in unison) [20]. This approach is also at the base of the Adler equation [21] and can be used to describe laser mode locking [22]. Figure 4 presents the results of a statistical study over a large region of zero exciton-photon detuning on the sample. The general behavior corresponds very well to what is expected for the two cases. The points appearing between the two limit cases of synchronized and desynchronized phase correspond to pairs of polaritons that do not feel the nonlinear interaction intensely enough to get synchronization. This transition can be described in the frame of the Adler

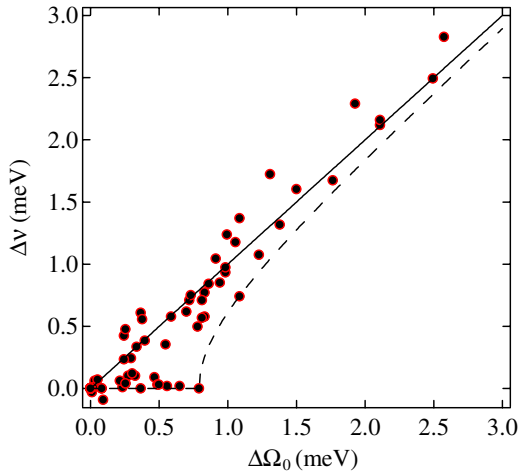


FIG. 4 (color online). Statistical study of the frequency detuning above threshold ($\Delta\nu$) as a function of the frequency detuning well below threshold ($\Delta\Omega_0$). Solid line: Nonsynchronized phase $\Delta\nu = \Delta\Omega_0$; the synchronized phase corresponds to $\Delta\nu = 0$. Dashed line: $\Delta\nu = (\Delta\Omega_0^2 - g^2)^{1/2}$, with $g = 850 \mu\text{eV}$.

equation, where only a nonlinear coupling is taken into account. Considering two coupled oscillators with initial frequency detuning $\Delta\Omega_0$, the evolution of the relative phase $\psi(t)$ between them is $d\psi(t)/dt = \Delta\Omega_0 + g \sin[\psi(t)]$, with g the coupling constant. Synchronization occurs when $d\psi(t)/dt = 0$, which can be achieved only if g is greater than $\Delta\Omega_0$. The transition can thus be described with only one fitting parameter, the critical value of $\Delta\Omega_0$, below which synchronization occurs, that gives an upper bound for an effective polariton coupling constant (see Fig. 4). For a model specific to polariton condensation, we refer to [14], where a complete understanding of the conditions required to observe a synchronized phase beyond the Adler equations can be found. It has been also addressed in Ref. [23].

To conclude, in the present work we have characterized the formation of polariton condensate(s) in a disordered medium. Synchronization allows building up a single ground state extended over a region larger than the typical disorder correlation length. From what we understand, the observation of synchronization rules out the interpretation of our results in terms of Bose glass, recently addressed for polaritons [24], where such extended coherence is not expected [25]. However, at some positions the disorder can destroy these correlations and produces independent condensates. In the case of polariton parametric scattering [26], the possibility of synchronization could be probed by interferometric measurements [27]. Focusing on a single potential well will allow us to minimize the influence of the disorder. This should prove useful for the characterization of the first excited modes and the investigation of the interactions in the condensate.

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