Dynamic exciton-polariton macroscopic coherent phases in a tunable dot lattice

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We report on the enhancement of the nonlinear properties of exciton-polariton macroscopic coherent phases created by resonant optical excitation under the spatial modulation of a square acoustic lattice. The confinement increases the local particle density by preventing particle drift due to repulsive interactions and modifies the coupling to the exciting laser field. Both effects lead to the enhancement of the nonlinear properties of the system, which is reflected in a reduction of the pump laser intensity threshold of formation of the coherent phase, pronounced blueshift of the emission energy and screening of the acoustic potential.

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Bosonic particles undergo a phase transition to a macroscopic quantum phase (MCP) at a critical particle density. In the solid state, MCPs can be formed by exciton polaritons, quasiparticles resulting from the strong coupling between photons and excitons in a semiconductor microcavity. The polariton MCP is a nonequilibrium state due to photon losses and can be maintained either by resonant laser excitation or by relaxation from an nonresonantly excited exciton reservoir. Independently of the excitation process, polariton MCPs exhibit spontaneous symmetry breaking similar to the phase transition of a thermal equilibrium Bose-Einstein condensation (BEC) as well as the characteristic properties of a system possessing a macroscopic wave function, such as extended spatial and temporal coherence. Importantly, polaritons possess strong nonlinear properties arising from the repulsive interaction between the exciton dipoles. Exciting discoveries related to interactions in resonantly excited polaritons MCPs have been reported, including superfluid-like and solitonic behavior as well as vortex imprinting.

Placing a MCP in a tunable periodical potential with period smaller than the size of the MCP has remarkable consequences, as demonstrated by experiments on atomic BECs in optical lattices. The periodic modulation creates a lattice of minicodensates while the potential tunability provides the control of the intersite interactions required to explore effects associated with the Bose-Hubbard model including Mott transition and particle blockade. For polaritons, different techniques have been developed to create confinement potentials for polaritons. Their exploitation for the fabrication of polariton lattices of interacting MCPs has, so far, been restricted due to the limited (or missing) tunability of the potential or inherent technological problems related to microstructuring. Exceptions are microstructured wires, static arrays of zero-dimensional (0D) polariton BECs produced by optical confinement and employed to demonstrate condensation in high-order orbital states, as well as acoustic potentials introduced by our group as an approach for the formation of tunable arrays of 1D MCPs with controlled intersite interactions based on the modulation by an elastic wave.

In this work, we investigate the nonlinear properties of a uniform lattice of 0D polariton MCPs created via tunable acoustic modulation. In contrast to the 1D lattices addressed in Ref. 14, the tunable square lattices investigated here provide a framework to study the transition from an extended MCP to a new physical regime, where the full spatial confinement completely suppresses particle drift preventing the establishment of long-range spatial coherence. Spectroscopic images of the MCP lattice show that the increased local particle density \(N\) in this regime leads to energy blueshifts due to polariton-polariton interactions which are considerably larger than in 1D and 2D MCPs. In particular, the laser Gaussian intensity profile determines a local \(N\) value and, thus, the blueshift, which becomes the main factor determining the energy of the MCP lattice sites. Furthermore, the optical threshold intensities for the formation of the MCP reduce with the lattice amplitude, particularly for MCPs with high photonic fraction. The threshold reduction is attributed to two mechanisms. The first is the increased \(N\) due to confinement. The second is associated with the resonant excitation scheme based on optical parametric oscillation (OPO). We show that the modulation of the polariton dispersion by the lattice relaxes the momentum conservation requirements necessary for OPO. Finally, we show how the nonlinear interactions screen the acoustic potential. This is demonstrated by monitoring the energy difference between the MCPs at the potential minima (s-like states) and those at higher energy formed at lower symmetry states of the periodic potential (p-like states) over several lattice periods.

The samples consist of GaAs-based \(\lambda/2\) microcavities with three pairs of 15-nm-thick quantum wells (QWs) and a Rabi splitting \(\Omega = 6.8\) meV. We present results for two polariton samples with photonic fractions of 64% (photon-like) and 42% (exciton-like) (corresponding to energy detunings between the bare photon and exciton energies of \(-2\) and 1 meV, respectively, as measured by standard methods). The 170 × 170 \(\mu m^2\) sinusoidal acoustic lattice [cf. Fig. 1(a)] was formed by the interference of two nonpiezoelectric surface acoustic waves (SAWs) of wavelength \(\lambda_{SAW} = 8 \mu m\) propagating along two orthogonal \(\{100\}\) surface directions. The modulation of the polaritons by the SAW relies on changes on the excitonic band gap and the optical microcavity resonance. Optical excitation was carried out with a Gaussian laser beam (diameter 60 \(\mu m\)) quasiresonant with the lower polariton branch with...
variable incidence angle $\theta_L$ and intensity $P_L$. The light emitted by the sample was collected along the narrow rectangular slit displayed in Fig. 1(a). Spatially resolved measurements reveal the real space emission of the dot array. Angular resolution allows us to map the angle $\theta$ dependence of the emission energies, yielding the in-plane polariton dispersion (energy vs planar momentum $k_p = k \sin \theta_L$), the in-plane momentum of the laser photons is given by $k_L = (2\pi/\lambda_{\text{laser}}) \sin \theta_L$, where $\lambda_{\text{laser}}$ is the wavelength of the laser.

At low $P_L$ (i.e., well below the threshold $P_{\text{th}}$ of formation of the MCP), an uncorrelated polariton gas with broad energy and momentum distribution forms. The angular resolved photoluminescence (PL) measurement in Fig. 1(c) reveals the dispersion [$k_{\parallel}$ lies along the slit in Fig. 1(a)] of the lower polariton branch of the photon-like sample, which is parabolic for small $k_{\parallel}$, as shown by the red line fitted to the data. Figure 1(e) displays the dispersion under modulation by the SAWs. The 2D SAW potential folds the parabolic dispersion into a square mini-Brillouin-zone (MBZ) with dimensions $k_{\text{SAW}} \times k_{\text{SAW}}$ ($k_{\text{SAW}} = 2\pi/\lambda_{\text{SAW}} = 0.78 \mu m^{-1}$). Since the slit is oriented at 45$^\circ$ with respect to the propagation directions of the SAWs, the dispersion of the dot lattice is plotted along the diagonal of the first MBZ with magnitude $\sqrt{2}k_{\text{SAW}}$. The two lowest lying states have $s$- and $p$-like symmetry. The fact that their dispersion is essentially flat indicates that they are strongly localized in the SAW potential. The redshift relative to the unperturbed dispersion is attributed to the repulsive interactions with upper levels of the polariton dispersion introduced by the SAW. The red line is a fit (see Supplemental Material), from which we determine a potential modulation amplitude per SAW beam equal to $\Phi_{\text{SAW}} = 0.66 \text{ meV}$. The peak-to-peak potential modulation $4\Phi_{\text{SAW}} = 2.6 \text{ meV} > \Omega/3$. The potential has a saddle point which determines the barrier height $2\Phi_{\text{SAW}}$ between two neighboring minima [cf. Fig. 1(b)].

As shown in Fig. 1(a), the propagation paths of the dots are spatially separated along the slit by a distance of $\lambda_{\text{SAW}}/\sqrt{2} = 5.6 \mu m$. The paths appear as maxima in the time integrated and spatially resolved PL image in Fig. 1(d) (spatial resolution of $\sim 3 \mu m$) revealing the spatial distribution of the folded states of Fig. 1(e). The $s$ states in the first MBZ are located at the minima of the SAW potential. The $p$ states have nodes at the minima of the potential and maxima spatially shifted relative to those of the $s$ states (see Supplemental Material). Note that higher levels are also affected by the lattice.

Increasing the pump laser intensity $P_L$ induces the formation of a polariton MCP. In the resonant excitation regime three polariton MCPs form. These correspond to the well-known OPO signal ($s$), pump ($p$) and idler ($i$) states created: two pump polaritons are scattered into signal and idler states in a process which conserves energy $E_{i} = E_L$, $E_{p}$ and momentum $k_{0} = k_{p} + k_{i}$. Importantly, the total phase (i.e., the sum of the pump, idler, and signal phases) is conserved in the scattering process, but neither signal nor idler inherit their phase from the pump. Figure 2(a) displays the real space emission of an unconfined photon-like signal MCP (lowest energy, $k_{\parallel} = 0$) excited by a laser [i.e., the pump state, red dot in Fig. 1(a)] with $k_{0} = 1.7 \mu m^{-1}$ and $P_L$ slightly above the threshold $P_{\text{th}} = 100 \mu m$. The idler state at $k_{i} = 2k_{0}$ and higher energy was not registered in our experiments. The distinctive features of formation of a MCP are observed: single mode emission with reduced linewidth ($2.4 \mu m$), intensity increase of three orders of magnitude relative to the incoherent emission in Fig. 1, as well as a pronounced blueshift ($\Delta E_{\text{FS}}$) with respect to the bottom of the dispersion below threshold ($E_{\text{LOW}}$).

An unconfined polariton MCP has uniform emission energy regardless of its size. Under a Gaussian excitation spot the polaritons are expelled from the high density regions due to the repulsive polariton-polariton interactions, acquiring thus kinetic energy. These polaritons redistribute the energy and maintain it constant over the MCP ensuring the establishment of the spatial coherence. The diameter of the photon-like MCP in Fig. 2(a) is approx. $20 \mu m$ at threshold and increases by a factor of 2 when $P_L$ doubles [Fig. 2(b)] due the increase of the area with local optical flux exceeding $P_{\text{th}}$ and the lateral expulsion of particles. Unconfined exciton-like MCPs have a smaller diameter ($5 \mu m$) at threshold and also expand with $P_L$ (not shown). The sizes of polariton MCPs at threshold are expected to be smaller for exciton-like states due to their larger mass and broader linewidths.
under the SAW dot potential for \( \Delta E_{BS} \) and under the same illumination conditions and under the SAW dot potential for \( P_\ell = 90 \) mW. \( \Delta E_{BS} \) denotes the energy blueshift with respect to the \( s \) states of uncondensed polaritons with energy \( E_{LOW} \) (white dashed line). The vertical dotted lines are guides delimiting the MCP edges.

Under the SAW potential [Figs. 2(b) and 2(c)], a series of dot MCPs with size \( \lambda_{SAW}/2 = 4 \mu \text{m} \) (limited by the imaging resolution) and different \( \Delta E_{BS} \) are observed. This suggests a local increase of \( N \) resulting from the reduction of lateral polariton drift due to the confinement. Dot-like emission is also observed for pump polaritons (not shown), thus proving that the acoustic field also modulates the pump states. In an OPO, \( \Delta E_{BS} \) is determined by interactions among the pump, signal, and idler MCPs. Considering that the linewidth (i.e., loss ratio) of the idler is larger than the one of the signal and that the pump particle density \( N_{pump} \) is much larger than those of the signal and idler, one can approximate \( \Delta E_{BS} \sim g N_{pump} \), where \( g \) is an effective interaction constant proportional to the excitonic fraction of the polaritons. Since the acoustically induced changes in the dispersion are small, this expression for \( \Delta E_{BS} \) is expected to be valid under a SAW. We therefore attribute the enhanced blueshift to a local increase of \( N \) since the polaritons cannot drift away from the dot region. The difference in the energy of the MCP dots across the Gaussian excitation spot is given thus by the potential energy under different local \( N \), frustrating the establishment of coherence over dimensions larger than the dot size. The spatial dependence of \( \Delta E_{BS} \) then follows the Gaussian intensity profile of the laser. Note also that the dot MCP region is always larger than the one for the unconfined MCP (indicated by the white dotted lines), which indicates a lower threshold, as discussed in detail below.

Figures 3(a) and 3(b) show the full dependence of \( \Delta E_{BS} \) on the pump laser intensity \( P_\ell \) for photon- and exciton-like MCPs, respectively. For the dots (white circles), only \( \Delta E_{BS} \) for the central dot is plotted. Notice that \( \Delta E_{BS} \) is always larger for the dots, which reflects the larger value of \( N \). \( \Delta E_{BS} \) increases rapidly at low \( P_\ell \) values, and then tends to saturation when no more pump polaritons can be injected into the cavity. For both photon- and exciton-like MCPs, the fact that at a fixed \( P_\ell \) \((100 \text{ mW, for example}) \) \( \Delta E_{BS} \) is \( \sim 1.4 \) times larger for the dots indicates an increase in \( N \) by a factor of \( \sim 1.4 \) relative to the unconfined case. This means that local losses resulting from the particle drift are reduced by approximately the same factor by the lateral confinement.

Further insight into the effects of confinement can be gained from the behavior of the threshold pump laser intensity \( P_{th} \). In Figs. 3(a) and 3(b), the point at the lowest \( P_\ell \) indicates \( P_{th} \) in each case. We first discuss the photon-like MCPs where, interestingly, \( P_{th} \) for the dots is a factor of \( \sim 8 \) smaller than for unconfined MCPs. Control experiments\(^{22}\) show that 50% of the reduction in \( P_{th} \) can be attributed to effects of temperature induced by the rf excitation.\(^{23}\) In addition, the strong decrease in \( P_{th} \) cannot be accounted for only by the 1.4-fold reduction in local losses, thus suggesting the existence of a second mechanism for threshold reduction. The latter is assigned to the improved OPO phase matching under confinement,\(^{24}\) which creates flat dispersion branches [cf. Fig. 1(e)] and thus relaxes the pump angle requirements of the OPO process. The dependence of \( P_{th} \) on the laser incidence angle (i.e., \( k_L \)) gives...
Further evidence for the improved phase matching under an acoustic lattice. In the absence of it, photon-like MCPs could only be excited at \( k_L \simeq 1.7 \, \text{μm}^{-1} \). Under the SAW lattice, in contrast, the MCP can form with much lower \( P_{th} \) over a wide range of \( k_L \) [cf. Fig. 3(c)]\(^{25}\). The reduction in \( P_{th} \) explains why the dot MCP area extends beyond that of the unconfined MCPs in Fig. 2.

As shown in Fig. 3(b) (solid squares), \( P_{th} \) is lower for unperturbed exciton-like polaritons than for photon-like ones. This is attributed to the larger nonlinearity \( g \), less pronounced dispersion, and broader linewidths, which help to fulfill the OPO phase-matching conditions. Also, cavity losses are more easily compensated for due to the lower photonic fraction\(^{23}\). The decrease in \( P_{th} \) through the reduction of lateral drift induced losses by the confinement is negligible (open circles), which is attributed to the fact that, at threshold, the exciton-like MCP is not much larger than the dots (5 μm). The use of smaller \( \lambda_{\text{SAW}} \), however, should have a larger effect. The effects of confinement at larger \( N \) are, however, clearly visible, as the blueshift is larger for the dots at any \( P_{th} \). Notice that \( P_{th} \) and \( \Delta E_{\text{BS}} \) for the photon-like dot MCPs reach values comparable to those of the unconfined exciton-like MCPs with larger nonlinearity, attesting to the strong increase of their nonlinear properties.

The reduction of \( P_{th} \) for exciton-like MCPs through improved phase matching can be observed when the laser \( k_L < 1.5 \, \text{μm}^{-1} \) (the optimum resonance condition), as shown in Fig. 3(d) (filled squares). In contrast, for \( k_L = 1.5 \, \text{μm}^{-1} \), \( P_{th} \) increases. The latter is attributed to the modulation of the pump levels, which worsens the coupling to the pump laser. In addition, temperature effects which increase \( P_{th} \) in exciton-like polaritons are expected to become more important at high \( k_L \)\(^{23}\).

The previous results relate to \( s \)-like MCPs from the lowest branch of Fig. 1(c). By changing the excitation conditions, it is also possible to observe \( p \)-like MCPs, as shown in Fig. 4(a). The excitation process for these MCPs is presently not completely understood. It may involve scattering into the strongly confined \( p \) states from an incoherent exciton reservoir\(^{25,26}\) or directly from the pump through additional scattering channels created by the periodic modulation\(^{27}\).

Interestingly, the spatial variation of the \( p \)-state energy levels \( E_p \) across the Gaussian excitation spot is less pronounced than for the \( s \) states. This results from the fact that the \( p \) states are spatially separated from the high density of pump and \( s \) states.

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13 J. Bloch et al., private communication (unpublished).
22 To minimize thermal loading, both the laser and the rf power were applied in the form of short pulses (2 ms) with a 10% duty cycle.
25 For photon-like polaritons, the modifications of the polariton Hopfield coefficients by the SAW strain field may also contribute to the threshold reduction. The changes, however, are small in this case (~10%) and can be neglected.