Phase diagram of microcavity exciton-polariton condensates

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Abstract – In this work, we study the exciton-polariton condensate phase transition in a microcavity matter-light system in which electron-hole Coulomb interaction and matter-light coupling effects are treated on an equal footing. In the framework of the unrestricted Hartree-Fock approximation applying the two-dimensional exciton-polariton model, we derive the self-consistent equations determining simultaneously the excitonic and the photonic condensate order parameters. In the thermal-equilibrium limit, we find a condensed state of the exciton-polariton systems and phase diagrams are then constructed. At a given low temperature, the condensate by its nature shows a crossover from an excitonic to a polaritonic and finally photonic condensed state as the excitation density increases at large detuning. Without the detuning, the excitonic condensed state disappears whereas the polaritonic or photonic phases dominate. The crossover is also found by lowering the Coulomb interaction at a finite matter-light coupling. Lowering the Coulomb interaction or increasing the temperature, the excitonic Mott transition occurs, at which the exciton-polariton condensates dissociate to free electron-hole/photon. Depending on temperature and excitation density, the phase transition of the exciton-polariton condensates is also addressed in signatures of photoluminescence mapping to the photonic momentum distribution.

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Introduction. – Transitions to quantum condensed phases, specially Bose-Einstein condensation (BEC), in solid-state systems have stimulated a great research effort for several decades [1,2]. In the condensed state, pure quantum effects can be observed at a macroscopic scale [3]. Above a critical density, bosons can undergo BEC if the temperature is small enough. The so-called-BEC transition temperature is inversely proportional to the mass of the boson [2]. Finding a boson with small effective mass is thus a crucial point in raising the transition temperature. The exciton —a bound state formed by a Coulomb correlated electron-hole pair— is one of the light mass Bose particles [2]. The possibility of excitons condensing into the macroscopic phase-coherent ground state was theoretically proposed about 50 years ago [4,5]. In experiments, however, higher excitonic density enhances the exciton-exciton annihilation processes and a sufficiently high excitonic density is thus difficult to prepare and an excitonic BEC in a bulk crystal is therefore rarely established [6].

An applicable way to explore excitonic BEC is the use of a semiconductor quantum well embedded in an optical cavity [3,7]. In that so-called semiconductor microcavity, photons are prevented from escaping and cavity polaritons (bound states of excitons and photons) are therefore formed with long lifetime [8]. Moreover, at zero momentum, the cavity polariton has an extremely small effective mass and temperature and density criteria for establishing the BEC condensation are thereby practicably realizable in experiments. Indeed, a condensation of the cavity polaritons at room temperature has been observed [9].

In general, the exciton-polariton system is in non-equilibrium [10], however, in constructing the phase diagrams, we assume that the exciton-polariton system in a microcavity is in thermal equilibrium. This holds if the quasiparticles have a lifetime much longer than their thermalization time. In the case of positive detuning, it has been both theoretically and experimentally verified that the polariton gas in a microcavity can be well described by the thermodynamic BEC theory [11]. Recently, a polariton gas trapped in a high-$Q$ microcavity structure which is found in thermal equilibrium has been
created [12]. The thermal equilibrium may be considered as the limiting case of a non-equilibrium situation. In this case, the decay rates for the loss of cavity photons and of fermions, for instance due to phonons or impurities, into external bath variables become small [13].

To model a thermal-equilibrium coupled electron-hole/photon system in a microcavity, we use a many-body Hamiltonian. The electron-hole Coulomb interaction and an electron-hole pair coupling to the light field are treated on an equal footing. The ground state, i.e., at zero temperature, of the thermal-equilibrium microcavity exciton-polariton systems has been intensively studied and crossovers of excitonic-polaritonic and polaritonic-photon condensates have also been discussed [14–16]. Without photons, finite-temperature effects on the excitonic condensate have been addressed [17,18]. However, in these studies, all quantum fluctuations are neglected. That mean-field treatment, for instance, can be corrected by a projector-based renormalization method allowing to incorporate fluctuation processes [19,20]. However, due to the calculation being cumbersome, the application of the method to the polariton problem is limited to the one-dimensional situation only [16]. That is not the case of the problem in a microcavity which is considered a 2D system [21]. In our work, we study the Hamiltonian in the 2D case but limited to the unrestricted Hartree-Fock (UHF) approximation. Although depleting all fluctuations, the UHF approximation describes quite well, at least qualitatively, the quantum phase transitions in strongly correlated electron systems [22]. For positive detuning and treating the Coulomb interaction on an equal footing with the light-matter coupling, the temperature effects on the excitonic and photonic contribution to the polariton condensation have been investigated. The transition temperature of the polariton BEC is determined and then phase diagrams of the condensation states are constructed. Crossovers of the excitonic-polaritonic and polaritonic-photon condensate transitions are characterized by a change of a photonic condensate fraction value [14]. The exciton-photon fraction, in particular, is extremely relevant to analyze the stability of the polariton systems [23]. To connect with the experimental study in photoluminescence detecting the exciton-polariton condensations in the system, we also discuss the momentum distribution of photons depending on temperature and excitation density.

The theoretical approach. – To discuss the exciton-polariton system in a semiconductor microcavity, in what follows, we study a two-dimensional interacting electron-hole-photon model. In momentum space, its Hamiltonian is written as

$$\mathcal{H} = \sum_{k} e_k^e \phi_k^e + \sum_{k} e_k^h \phi_k^h + \sum_{q} \omega_q \psi_q^\dagger \psi_q$$

$$- \frac{U}{N} \sum_{k_1,k_2} e_{k_1+k_2}^e \phi_{k_1}^e h_{k_2}^h$$

$$- \frac{g}{\sqrt{N}} \sum_{kq} \left( \phi_{k+q}^e h_{k}^h + \text{H.c.} \right),$$

(1)

where $e_k^e$, $h_k^h$, and $\psi_k$ are the spinless electron, hole, and photon annihilation (creation) operators at momentum $k$, respectively. $N$ is a number of lattice sites. The first two terms in the Hamiltonian indicate a non-interacting part of the electron-hole system, where

$$e_k^e = -2t(\cos k_x + \cos k_y) + \frac{E_g + 8t - \mu}{2},$$

(2)

are the tight-binding dispersions of 2D free electrons and holes in the hypercubic lattice. In (2), $t$ denotes the particle transfer amplitude, $E_g$ gives the minimum distance (gap) between the bare electron and hole bands. Note that a semimetallic setting occurs when $E_g < 0$ and vice versa $E_g > 0$ establishes a semiconductor situation.

The third term in eq. (1) indicates the free-photon part with an excitation energy

$$\omega_q = \sqrt{(c q)^2 + \omega_e^2} - \mu.$$  

(3)

Here, $\omega_e$ is a zero-point cavity frequency and $c$ is the speed of light in the microcavity. In eqs. (2) and (3), $\mu$ is a chemical potential which was included to control the total number of excitations

$$n = \frac{1}{N} \sum_{q} \langle \psi_q^\dagger \psi_q \rangle + \frac{1}{2N} \sum_{k} \langle \phi_k^e \phi_k^e \rangle + \langle h_k^h h_k^h \rangle.$$  

(4)

The last two terms in Hamiltonian (1) are a local electron-hole Coulomb interaction and a local exciton-photon interaction, respectively. In principle, additional electron-electron and hole-hole Coulomb interactions might have been taken into account in the Hamiltonian. However, we leave these out here because they only lead to mere shifts in the one-particle dispersions $e_k^e$ and $e_k^h$. The model written in the Hamiltonian (1) is similar to the models of two hyperfine spin states close to a Feshbach resonance, discussing the context of BEC-BCS crossover in a degenerate Fermi gas [24]. By including fluctuations of the particle-particle interaction, the finite-temperature superfluid phase diagram in the latter models has been also addressed [25].

Apparently, the influence of $\mathcal{H}_{e-ph}$ in the Hamiltonian (1) becomes more important when the excitation energy of a particle-hole pair roughly agrees with a photon excitation. Therefore, for a later interpretation of
this effect one best introduces the so-called detuning parameter [14]
\[ d = \omega_c - E_g. \]  
(5)

Our study is limited to a positive value of the detuning parameter. In this situation, the polariton gas in the microcavity can be well considered in thermal equilibrium [11].

To address the formation of the exciton-polariton condensates in the electron-hole-photon system, we look for non-vanishing values of excitonic expectation and polarized photonic field indicating a kind of spontaneous symmetry breaking due to the coupling of excitons and photons [16,26]. The task can proceed if we adapt the unrestricted Hartree-Fock (UHF) approximation [27]. The UHF approximation allows decoupling with respect to the off-diagonal expectation values. Leaving out all fluctuation parts, an effective UHF Hamiltonian driving from eq. (1) reads
\[ \mathcal{H}_{\text{UHF}} = \sum_k \varepsilon^e_k \psi^e_k \psi_k + \sum_k \varepsilon^h_k \psi^h_k \psi_k + \Delta \sum_k (\psi^e_k \psi^h_k + \text{H.c.}) + \sum_q \omega_q \psi^q \psi^q + (\sqrt{N} \Gamma \psi^0_0 + \text{H.c.}), \]  
(6)

where all additional constants have been neglected. In eq. (6) the electronic excitation energies have acquired Hartree shifts
\[ \varepsilon^e_k = \varepsilon^e_k - \frac{U}{N} \sum_q \langle h^+_q h_q \rangle, \]  
(7)
\[ \varepsilon^h_k = \varepsilon^h_k - \frac{U}{N} \sum_q \langle e^+_q e_q \rangle, \]  
(8)

and additional fields
\[ \Delta = -\frac{g}{\sqrt{N}} \langle \psi^0_0 \rangle - \frac{U}{N} \sum_k d_k, \]  
(9)
\[ \Gamma = -\frac{g}{N} \sum_k d_k, \]  
(10)

where
\[ d_k = \langle e^+_k h^+_k \rangle = \langle s_{-k} c_k \rangle = d^*_k \]  
(11)

play a role of order parameters for the exciton-polariton condensates. In the exciton-polariton condensates, both the excitonic order parameter \( d_k \) and the photonic polarization \( \langle \psi^0_0 \rangle \) are non-zero. In this situation, we have restricted ourselves to the direct electron-hole pair coupling. An indirect case can be extended for a further study. \( \Delta \) in eq. (9) is the generalized Rabi frequency which represents the coherence of the system. Note here that both electron-hole and exciton-photon interactions make contributions to \( \Delta \), where their mutual influence in the formation of a condensate will be of interest. On the other hand, the shift \( \Gamma \) in eq. (10) leads to a polarization of the photonic subsystem. In case the detuning parameter \( d \) (eq. (5)) is small, the tendency for the formation of a photonic condensate is expected to be enhanced. In contrast, for large \( d \) the photonic contribution to \( \Delta \) should be small, at least for a not too large excitation density.

The non-interacting Hamiltonian form in eq. (6) can be simply diagonalized, then the expectation values are self-consistently evaluated [26]. In this way, we can determine both the excitonic and the photonic order parameters separately. Complexed phase structure of the exciton-polariton condensates depending on the model parameters is thus addressed.

**Numerical results.** – The UHF approach outlined in the previous section must be numerically evaluated of course. In doing so, we work in momentum space, on a discrete set of \( N = 200 \times 200 \) points. The solution of the whole self-consistent calculation is assumed to be achieved if all quantities are determined with a relative error less than \( 10^{-5} \). In the numerical calculation, all energies are given in units of the particle transfer amplitude \( t \).

Without loss of generality, we choose the cavity frequency \( \omega_c = 0.5 \). The physical scenario is not much different unless the cavity frequency is larger than the width of the bare band structure [16]. The latter case will be left to a future study. Assuming a thermal-equilibrium situation [8,14], in the calculation below, we restrict ourselves to a positive detuning [28].

To analyze the complex phase structure of the quantum condensed states, we separately determine two contributions to the polaritonic condensate order parameter \( \Delta \) on the right-hand side of eq. (9): \( \Delta_X = -\frac{g}{N} \sum_k d_k \) (excitonic condensate order parameter) and \( \Delta_{ph} = -\frac{g}{\sqrt{N}} \langle \psi^0_0 \rangle \) (photonic condensate order parameter). Step by step, we analyze the temperature dependence of both the contributions for varying excitation density and Coulomb interaction. Then the critical temperatures of the phase transitions are determined, and, as a consequence, the phase diagram of the condensates is constructed. Crossovers of the excitonic-polaritonic and polaritonic-photonic condensate transitions are characterized by a change of a photonic condensate fraction value (\( \Delta_{ph}/\Delta \)). If the fraction is in the range 20%–80% the system stabilizes in a polariton condensate [14]. Otherwise, the system stabilizes in a photonic condensate if the fraction is larger than 80% and an excitonic condensate if it is smaller than 20%.

**Excitation density dependence.** Firstly, we discuss the phase structures of the exciton-polariton system depending on the excitation density and temperature. In theory, temperature and density are the parameters driving the phase transition. The excitation density injecting the polariton density is an easily tunable parameter, and so it is often chosen as the experimental control parameter [3]. Figure 1 shows the excitonic (\( \Delta_X \)) and photonic (\( \Delta_{ph} \)) condensate order parameters as functions of temperature for some values of the excitation density \( n \) at detuning \( d = 4 \), matter-light coupling \( g = 0.2 \), and Coulomb interaction \( U = 2 \). At small excitation density \( n = 0.2 \),
Panel (a)), one finds a domination of the excitonic condensate order parameter in comparison to the photonic condensate order parameter in the entire temperature range. In this situation, the minimum of the photonic band is far from the chemical potential [16,26]. The photonic condensate order parameter is therefore small and vice versa the excitonic condensate order parameter is enlarged. Phase-space (Pauli-blocking) effects become more important and the condensate typifies the excitonic coherent state, the light component is negligible. Increasing the excitation density, overlap of the conduction and the hole bands develops, the possibility of electrons and holes coupling to form an excitonic bound state increases. In this case, the chemical potential also comes closer to the minimum of the photonic band and photonic effects come into play. Both excitonic and photonic condensate order parameters are intimately connected indicating a polaritonic condensation. As a result, the bound exciton-polariton state in a microcavity structure develops (fig. 1(b)). The condensate turns from an excitonic to a polaritonic condensed state. Increasing the density further, at low temperature, the photonic condensate order parameter continues its increase, whereas the excitonic condensate component saturates (fig. 1(c), (d)). As a consequence, the system can be classified as a photonic condensate. In contrast, at temperatures close to $T_c$, the excitonic condensate order parameter becomes comparable to the photonic condensate component. Increasing temperature, this displays a crossover to the polaritonic from the photonically dominated wave function. At large excitation density (fig. 1(d)), one finds the polaritically dominated wave function in the entire temperature range.

Here, for all excitation densities, the excitonic and photonic condensate order parameters decrease monotonically with increasing temperature. They completely disappear simultaneously if the temperature is larger than a critical value $T_c$. At $T > T_c$ all exciton-polariton bound states are deformed and the system is in the normal liquid state. $T_c$, therefore stands for the exciton-polariton condensate transition temperature. For a given set of $d$, $U$, and $g$, fig. 1 also shows us that the critical temperature increases if the excitation density increases.

The phase diagram summarizing the exciton-polariton condensates in the $T$-$n$ plane for weak and strong detuning $d$ at $U = 2$ and $g = 0.2$ is illustrated in fig. 2. At weak detuning ($d = 0$, panel (a)) one finds $E_g = 0.5$, which leads the system to a semiconducting bare band structure [14,16]. Because the photonic frequency is small ($\omega_0 = 0.5$), the chemical potential rapidly reaches the photonic minimum energy and there are mainly photonic excitations in this case. The excitonic condensate is not found, and instead, at low temperature one finds the polariton condensate and then the photonic condensate if $n > 0.2$. In contrast, at strong detuning we enter the semimetallic situation. If the excitation density is small, the minimum of the photonic band is far from the chemical potential. In this situation, the photonic condensate order parameter is therefore small and the excitonic condensate part is enlarged. The phase diagram in this case is shown in fig. 2(b). Here, one finds the excitonic condensation state at low excitation density. The polaritonic and then the photonic condensates appear if the excitation density gradually increases. Moreover, at strong detuning, depending on temperature one also finds polaritonic-excitonic (at low excitation densities) or photonic-polaritonic (at high excitation densities) condensate crossovers, whereas, at zero detuning, only the latter is found. In both cases of the detuning, increasing the excitation density always enhances the condensate transition temperature $T_c$.

**Coulomb interaction dependence.** Next, we analyze the phase structure of the exciton-polariton system depending on the Coulomb interaction. Figure 3 shows the excitonic ($\Delta_X$) and photonic ($\Delta_{ph}$) condensate order parameters as functions of temperature for some different
values of the Coulomb interaction at \( g = 0.2, d = 2, \) and \( n = 0.2. \) As a starting point, we address ground states of the system, i.e., at \( T = 0. \) When the Coulomb interaction is extremely low \( (U = 0.1, \text{ panel (a)}), \) electrons and holes are not able to couple with each other to create pairs, the exciton thus does not exist \([20]\). In this case, one finds only the photonic condensate due to the matter-light coupling. At an intermediate Coulomb interaction, both \( \Delta_X \) and \( \Delta_{ph} \) are comparable and the system stabilizes in the polariton condensate (see fig. 3(b) and (c)). Increasing the Coulomb interaction, the excitonic condensate order parameter increases, whereas, the photonic part decreases. At \( U = 4 \) (fig. 3(d)) the excitonic condensate order parameter is significantly dominant to the photonic one. The system thus settles in the excitonic condensation state. Increasing temperature, similar to the results addressed in fig. 1, both \( \Delta_X \) and \( \Delta_{ph} \) monotonically decrease and then simultaneously disappear at a critical value. This transition temperature \( T_c \) increases if the Coulomb interaction is increased.

In fig. 4, we show phase diagrams of the exciton-polariton condensates in the \( T-U \) plane for some values of matter-light coupling \( g \) at detuning \( d = 2 \) and excitation density \( n = 0.2. \) In the case of no matter-light interaction \((g = 0, \text{ panel (a)}), \) the system is in a quantum condensed state only if the Coulomb interaction is large enough. Due to the electron-hole coupling only, the system settles in the purely excitonic condensation. The critical temperature of the transition increases with increasing Coulomb interaction \([20]\). When the temperature is higher than the transition temperature, the electron-hole pairs are unbound and the system settles in the electron-hole plasma with free-photon states. One also obtains this feature by lowering the Coulomb interaction at a given temperature. This excitonic-Mott transition has been extensively studied both theoretically and experimentally for the exciton-polariton systems \([29,30]\). Enlarging the matter-light coupling, low-temperature bound states of excitons and photons can be formed and the polaritonic condensate appears. In this case, at a given low temperature, one finds both the photonic-polaritonic (at small \( U \)) and polaritonic-excitonic (at large \( U \)) condensate crossovers as discussed in fig. 2. Similarly to the phase diagrams illustrated in fig. 2, fig. 4 also displays the photonic-polaritonic and polaritonic-excitonic condensate crossovers depending on temperature.

**Momentum distribution of photons.** In experiments, the BEC state can be detected by measuring the angle-resolved photoluminescence (PL) intensity. A sharp peak in the PL signatures indicates large numbers of the states occupying the same energy, reducing a sharp peak in the momentum distribution. In exciton-polariton systems, it is convenient to map the PL signatures to the momentum distribution of photons \([31]\).

We first analyze the momentum distribution of photons as a function of the excitation density \( n \) at a given temperature. Figure 5 shows the magnitude of the momentum distribution for some values of \( n \) at \( T = 0.2, U = 2, g = 0.2, \) and \( d = 0. \) At low excitation density (left panel), it exhibits a smooth and low photonic distribution around \( k = 0. \) Increasing the excitation density, the distribution of photons at zero momentum develops (middle panel) and then forms a sharp peak (right panel) if the excitation density is higher than a critical value of the polaritonic BEC transition (see fig. 2).
peak in the momentum distribution appears due to the long-ranged phase coherence in the condensate state. At a given small temperature, when the excitation density is large enough, the coherence length exceeds the wavelength of the emitted radiation, the system establishes the condensate [3].

At a given excitation density, we continue discussing the momentum distribution of photons depending on temperature. Figure 6 displays the momentum distribution of photons at $n = 0.4$ for some different temperatures $T$ (other parameters are the same as in fig. 5). Similarly to the result analyzed in fig. 5, at low temperature $T < T_c$ (left and middle panels) the distribution shows a sharp peak at zero momentum, indicating the condensation state. Note here that the peak in the distribution appears due to the presence of phase textures in the condensate. Increasing temperature, thermal fluctuations develop and destroy the phase textures [32]. At high enough temperature ($T = 0.3$, right panel), the thermal fluctuations completely deplete the phase coherence. The distribution at zero momentum is therefore strongly smeared out and suppressed, the system is out of the condensation state. Moreover, from right to left, fig. 6 shows that the width of the momentum distribution shrinks with lowering temperature, and below the transition temperature, the emission mainly comes from the lowest-energy state at zero momentum. This scenario has been shown with increasing excitation density at a given low temperature in fig. 5 [3]. Due to the strongly electronic and photonic correlations, the system favors the exciton-polariton condensation states if the temperature is low and the excitation density is high enough.

**Summary.** – To summarize, in this paper we have developed the unrestricted Hartree-Fock approximation to analyze the phase diagram of the exciton-polariton condensate in a two-dimensional microcavity. Treating the electron-hole Coulomb interaction and matter-light coupling effects on an equal footing, the popular exciton-polariton Hamiltonian describing the polariton in a microcavity has been solved. We then derived self-consistent equations to determine simultaneously the excitonic and photonic condensate order parameters. In the thermal equilibrium limit, phase diagrams summarizing the exciton-polariton condensates in the $T$-$n$ plane for different detuning or in the $T$-$U$ plane for different matter-light coupling have been constructed. In both cases, the critical temperature monotonically decreases when decreasing the excitation density or the Coulomb interaction. Depending on the excitation density or the Coulomb interaction, the phase diagrams show both the excitonic-polaritonic and the polaritonic-photonic condensate crossovers. The phase diagrams also illustrate the excitonic Mott transition that the exciton-polariton condensates dissociate to a free electron-hole/photon if the temperature is high enough or the Coulomb interaction or excitation density are small enough. To connect with the experimental photoluminescence detecting the exciton-polariton condensations in the system, we also discussed the momentum distribution of photons depending on temperature and excitation density. If the temperature is lower and the excitation density is larger than the respective critical values, the system favors the exciton-polariton condensation states due to the strongly electronic and photonic correlations, indicated by a sharp peak at zero momentum in the momentum distribution of photons. These findings are promising for the development of the so-called polariton laser and Bose condensation at increased temperatures with wider-bandgap semiconductors such as ZnO or GaN [3].

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