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Nonclassical polariton phenomena in a triple-micropillar system

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We investigate quantum phenomena in a system of three coupled microcavities each supporting regime of strong coupling with exciton resonance. The possibility of observing a polariton blockade in a dimer and trimer configuration is discussed. We identify the resonance conditions corresponding to the onset of the photon blockade. The discovered quantum effects allow for using these systems as versatile sources of individual polaritons. Various manifestations of the quantum blockade can be tuned with the use of the pumping laser frequency. We demonstrate that the presence of the complex coupling between the pillars enhances manifestations of sub-Poissonian statistics of polaritons.

I. INTRODUCTION

Studying the quantum statistical properties of the non-linear light-matter systems is of a great interest. The harvest of which is a highly desirable for quantum information [1, 2], or for the creation of single-photon sources [3–5]. Quantum anticorrelations of the radiation emitted by the nonlinear systems typically stems from the spectrum anharmonicity [5, 6]. The anharmonicity effect leads to the fact that the first excited state of an oscillator is not resonant with the second excited state. So that, the appearance of two photons at the same frequency is unlikely in the system (but becomes possible in the presence of the dissipation-induced broadening). This is an effect of photon blockade [5, 7, 9–11] which allows for converting a coherent laser light into a stream of individual photons prepaired in the Fock state [12]. Typically, this phenomenon occurs in the low-dimensional systems where strong quantization decouples ground state from the higher energy levels. [12, 14–17]. Relatively strong interparticle interactions (nonlinearities) are also required [7, 15–20]. The mathematical ability for such suppression and separation of the flow into separate corpuscles is determined by the second-order correlation function \( g^{(2)} \).

Usually, a pronounced photon blockade effect requires a nonlinearity comparable to the losses in the system. the parameter \( g^{(2)} \) must not exceed 0.01 [8]. Unconventional quantum statistics implies weakly nonlinear systems (\( U < \gamma \)), which is a more reality a value of a nonlinearity in the condensed matter physics [7, 20–22].

To maintain stationary modes, resonators are usually used. A typical system for study interaction light and matter is low-dimensional systems - quantum wells placed in a microcavity [23–25] or based on quantum dots [23, 25]. We considered the exciton-polariton system formed in 0D pillar microcavities (MP)[26–29] with typical for this system weakly nonlinearity [30] \( U \approx 0.01\gamma \). Quantum statistical properties of the microcavity emission had been studied before [31, 32]. Namely, the antibunching, bunching and giant bunching phenomena have been discovered.

The Sub-Poisson statistics of radiation from MPs, including those supporting strong coupling between exciton and photon modes, was predicted theoretically in weak pumping [31] and for strong pumping [32]. However, the quantum nature of the polaritons themselves remains coherent in this case. The mechanism of photon blockade in the first case can be similarly explained by unconventional blockade. In the second case, the photon blockade mechanism is based on the asymmetry between the population of the photon and exciton modes, Poisson fluctuations of the exciton mode lead to strong super-Poisson fluctuations of the photon mode or anticorrelations effects with sub-Poissonian statistics, depending on the resonance frequency parameters of the system.

The article has the following structure: in the first section, a general model of a polariton trimer see 1c consisted of three closely placed MPs is introduced. The main kinetic equation for the density matrix of a cascade open quantum system is written. In the second section, we first consider the statistics of the polariton dimer, and then move on to the section devoted to the polariton trimer. The various types of coupling between MPs are considered.

II. MODEL

A polariton trimer is described by the following Hamiltonian written in the rotating wave approximation:

\[
\hat{H} = \hat{a}^{\dagger} \hat{G} \hat{a} + \sum_i \sum_{j \neq i} F_i (\hat{a}_i^{\dagger} + \hat{a}_i) \hat{a}_j + \sum_i U_i \hat{a}_i^{\dagger} \hat{a}_i \hat{a}_i^{\dagger} \hat{a}_i,
\]  

(1)

Here \( \hat{a} \) = \((\hat{a}_1, \hat{a}_2, \hat{a}_3)^T \) with \( \hat{a}_i \) being a polariton annihilation operator in the \( i \)th MP while \( U_i \) quantifies the strength of the interparticle interaction inside the \( i \)th MP [34]. The linear part of the Hamiltonian (1) is given by the matrix:

\[
\hat{G} = \begin{pmatrix}
\Delta_1 & g_{12} & g_{13} \\
g_{21} & \Delta_2 & g_{23} \\
g_{31} & g_{32} & \Delta_3
\end{pmatrix},
\]  

(2)

where \( \Delta_i = \omega_i - \omega_L \) is the detuning of the \( i \)th cavity from the frequency \( \omega_L \) of the coherent driving field \( F_i \) which
is applied to the $i$th MP, $g_{ij}$ is the coupling between $i$th and $j$th MPs.

We assume that only a second pillar is coherently pumped from outside, $F_{1,3} = 0, F_2 = \gamma_2$. The rest two cavities are populated due to the photon tunnelling from the pumped MP, which is described by the non-diagonal elements of $G$.

The Hamiltonian (1) is Hermitian provided that $G$ is Hermitian as well. Once this condition fails, this fact must be take in account in the govern equations for the quantum objects in order to preserve the normalization of either the wave function or the density matrix. We consider the general case of the matrix $G$. We have considered several cases: (I) Hermitian symmetric coupling; (II) broken coupling when $g_{13} = g_{31} = 0$; (III) we have introduced a non-reciprocal cascade coupling [35]. In the latter case, $g_{13} = ig_{c}$ and $g_{31} = 0$, where $g_{c}$ – a cascade interaction parameter [36], illustrated in Fig. 1a. (IV) we took two non-reciprocal cascade coupling which are shown in Fig. 1b with the parameters $g_{13} = \sigma + ig_{c}$ and $g_{31} = -ig_{c}$, where $\sigma$ – is a cascade interaction parameter with rotated relative phase of the annihilation operator $a_1$ [13]. The cascade coupling is realised in the presence of the strongly dissipative mediate system, which can be adiabatically excluded. In our case, this can be realised by the unidirectional waveguide [40] as it is schematically shown in Figs. 1a,b.

For the description of the open quantum system we use a general kinetic equation for the density matrix $\rho$ [41]:

$$\frac{d\rho}{dt} = -i\left[\hat{H}, \rho\right] + \gamma_1 D[a_1] + \gamma_2 D[a_2] + \gamma_3 D[a_3] + 2|g_c| D[a_1 + a_3], \quad (3)$$

where we define the dissipator super-operator $D[a] = a\hat{a} + \frac{1}{2}\{\hat{a}^+\hat{a}, \rho\}$ which describes interaction with the environment with rates $\gamma_1, \gamma_2, \gamma_3$. Here $\{\{\ldots\}\}$ stands the anticommutator. Last term in (2) describes the cascade coupling with the constant $g_c = \theta_{gain}\sqrt{\gamma_1\gamma_3}/2$, where $\theta_{gain}$ is the gain coefficient of the cascade interaction [36]. We can rewrite (3) in a matrix form and obtain linear differential equations on the density matrix elements

$$\rho = \sum_{n'^{m'}n',nml} \rho_{n'^{m'}n',nml} |n'n'\rangle \langle nml| \quad (4)$$

Here the basis states $|n'n'\rangle$ are the Fock states with $n$ (and $l$) photons occupying the first (second and third) MP. The corresponding differential equations for $\rho_{n'^{m'}n',nml}$ are given in the Appendix, see (7).

The manifestations of the nonclassical behaviour of the MPs system, we quantify by the second-order correlation function. For the first MP it can be defined as:

$$g_1^{(2)}(0) = \frac{\langle a_1^+a_1^+a_1a_1\rangle}{\langle a_1^+a_1\rangle^2} = \frac{\sum_{ij,k} \langle i(l-1)\rangle \rho_{ijkl}}{\langle a_1^+a_1\rangle^2} \quad . \quad (5)$$

And analogically for other MPs.

The coupled microcavities model in the frame of a lower-polariton truncated basis approximation [34] with parameters are used [33]: $g = 5.62\gamma, U_1 = U_2 = U_3 = U_{LP} = 0.01\gamma_1, \gamma_1 = \gamma_2 = \gamma_3 = \gamma, F_1 = 0, F_2 = \gamma_1, F_3 = 0, \hbar\gamma = 0.0274$ meV.

We solve the master equation (3) numerically. In particular, the qupit python library was employed with the Fock state space size truncated at $M = 3$. Using $M > 4$ in the trimer configuration requires extraordinary large computational resources when scanning over the wide range of parameters. We have check our results with $M = 5$ at several important limit cases and confirmed the validity of the used cut-off value in the blockade regime which is typically realised at the weak pumping.

III. RESULTS

A. A dimer

To start with, we review two simple schemes demonstrating photon blockade. In particular, we consider a single coherently pumped MP ($g_2 = g_3 = 0$) and the MPs dimer - the one of the MP is pumped by a coherent laser and coupled to the other MP via a tunnel coupling ($g_3 = g_{13} = g_{31} = 0$). Fig. 2 shows a comparison of the $g_1^{(2)}$-parameter for a single microcavity and for the coupled microcavities systems. In a weakly nonlinear system at $U < \gamma$, the sub-Poissonian effects are weakly

FIG. 1. (a) Scheme with cascade connection. Arrows are show to type coupling: double arrow is Josephson connection, red arrow is non-reciprocal cascading connection; (b) Scheme with double cascade connection. Here we multiply the cascade coupling constant by the phase factor $e^{i\phi}$, in given case $\phi = \pi/2$ [13]; (c) Settling polariton states in a trimer system.
manifested, \( g^{(2)} \sim 0.98 - 0.99 \) (compare with the results from Ref. [7]). For a dimer, one can observe sub-Poisson statistics in the region of weak nonlinearities where a single pillar demonstrates just a weakly non-classical behaviour. Stationary pumping of one of the micropillars (MPs) of the dimer leads to population of the ground state and tunneling of the photon into the neighboring resonator. The population of subsequent states is blocked by the destructive interference of quantum trajectories [22]. This lead to quantum blockade of the first pillar which is pumped. The effect of polariton blockade statistic is achieved at sufficiently weaker nonlinearity.

In addition to nonlinearity, an important parameter of the system is the detunings of the resonant frequency of the MPs from the pump frequency. At some detuning values, the effect of polariton blockade is observed. We varied the values of all detunings and obtained the minimum values of the parameters \( g^{(2)} \), with some optimal detuning value. The effect of photon (or our case polariton) blockade in the dimer is achieved at negative detuning \( \Delta_{1} < 0 \), [22], which is not typical for a blockade with a single microresonator [7]. The condition of optimal detuning parameters which minimize \( g^{(2)} \) is \( \Delta_{\text{opt}}/\gamma_{1} = \frac{1}{2}(\sqrt{(U_{1}/\gamma_{1})^{2} + 1} - (U_{1}/\gamma_{1})) \) with \( g^{(2)} = 1 - 2U_{1}/\gamma_{1} \Delta_{\text{opt}}/\gamma_{1} \) [7].

This means that the antibunching effect itself in the resonant fluorescence of \( \omega_{L} = \omega_{1} + (n_{1} - 1)U \). For the optimal detuning parameters, the \( g^{(2)} \) parameter reaches 0.0013 at the typical value of the strength of nonlinearity \( U_{1} = U_{2} = 0.01\gamma_{1} \), while for a single micropillar it does not descend below 0.98 at the same parameters.

A more pronounced manifestation of the sub-Poissonian statistics in a coupled micropillars determines the use of such a system for creation of sources of single polariton states. This further inspires studying quantum blockade in the more complex systems. Due to the unconventional blockade mechanism proposed in [22], one can choose such resonance conditions that the quantum trajectories of the population of the states with \( n_{1} > 1 \) will be suppressed or unlikely (in the weak pumping mode). However, it is worth noting that the statistics for the second MP is close to the Poisson one.

B. Polariton trimer

Let’s add one more MP between two MPs. In the case of a conventional (reciprocal) tunneling coupling between subsystems, the minimal value of the second order is around \( \sim 0.6 \) for any MP. In this case, there is no pronounced sub-Poisson statistics for polaritons. However, introducing nonreciprocity crucially alters statistical properties. Using the configuration shown in Fig. 1 with a cascade connection between the unpumped MPs allows achieving quantum blockade regime. The considered system demonstrates the effect of polariton blockade in the area of those parameters that are shown in Fig. 3 with \( \theta_{\text{gain}} = 2.5 \). The minimal value of the second order of the first micropillar connected according to the scheme in Fig. 1a is 0.0015. The analysis of the probability amplitudes and the correlation function parameters demonstrates the existence of the regions of a significant decrease in the probability of occupying of polaritons states with \( n_{1,3} > 1 \) in the first and the third MPs. These areas of quantum blockade have a characteristic shape for two different cases, see Figs. 3 and 4. The quantum blockade region in the case of correlations between MPs has the form of two hyperbolas, see Fig. 5b. The appearance of hyperbolas is due to a real non-reciprocal connection in as part of complex coupled.

Using the cascade connection scheme shown in Fig. 1a, we can obtain the sub-Poisson statistics of polaritons in the blockade regime, which is demonstrated in Fig. 3, similar to the dimer case, but for different MPs. But the MPs are not correlated, there is only a small frequency domain of \( \Delta_{1,3} \) parameters, where there are small correlations between MPs, see Fig 5a, while using a more complex double cascade connection allows the pillars to be correlated and the patterns of the resonant regions become identical, Fig. 4 and Fig. 5b.

Figure 5 demonstrates a cross-correlation function:

\[
g^{(2)}_{ij} = \frac{\langle a_{i}^\dagger a_{j}^\dagger a_{j} a_{i} \rangle}{\langle a_{i}^\dagger a_{i} \rangle \langle a_{j}^\dagger a_{j} \rangle} .
\]

If the correlator is close to zero, the subsystems are uncorrelated. However, if it is negative, the subsystems are anticorrelated, i.e. if the fluctuations increase in one system, they decrease in the other. If the correlator is greater than zero, then the fluctuations in the system are correlated.

Introducing the cascade connection allows for observation of the polariton blockade but at different values of
the detuning parameter. The manifestations of a blockade in different pillars are uncorrelated and appear at different parameters. The introduction of a double cascade connection makes it possible to correlate the effects of polariton blockade. As it can be seen from Fig. 5b, the white area inside the hyperbolas is the area of correlation between the first and third MPs.

**IV. DISCUSSION**

**Q1.** How does a complex non-reciprocal connection by multiple paths with different phases work?

**Q2.** How is such a non-Hermitian connection taken into account in the master equation so that the trace is preserved? \( \text{Tr}(\rho) = 1 \)

**Q3.** How is the internal microcavity statistics of exciton-polaritons measured?

FIG. 3. Polariton statistics in the trimmer scheme with a pure cascade coupling between the first and the third pillars. The \( g^2 \)-function in the first (a,b), second (c,d) and third (e,f) pillars as a function of the detunings \( \Delta_1 \) and \( \Delta_3 \) at the following parameters: \( U/\gamma = 0.01, g_{12} = g_{21} = |g_c| \) and \( g_{13} = ig_c, g_{31} = 0 \). The meaning of \( g_c \) is different in the paragraph next to Eq. 2.

FIG. 4. The second order correlation function of the first (a,b), second (c,d) and third (e,f) pillars for the lower-polaritons in a trimmer scheme as a function of detunings \( \Delta_1 \) and \( \Delta_3 \) at the following parameters: \( U/\gamma = 0.01, g_{12} = g_{21} = |g_c| \) and \( g_{13} = ig_c, g_{31} = 0 \). The other parameters are given in text.

FIG. 5. (a) the correlation function between the first MP and third MP, (c) the second and third MPs, (d) the first and third MPs in case simple cascade coupled for parameters the same as on the fig 3, (b) the correlation function between the first MP and third MP, in case complex cascade coupled parameters take from case on the Fig. 4.
V. REMAINS TO DO

Do separate of the type coupling by absaz and implementery in trimer chapter. Namely: (I) Hermitian symmetric coupling; (II) broken coupling when $g_{31} = g_{31} = 0$; (III) non-reciprocal cascade coupling and (IV) double non-reciprocal cascade bond with the parameters $g_{13} = ig_{e}$ and $g_{11} = -ig_{e}$. Explain the physical implementation of the last case. Display the physical implementation of the couple in case in Figure 1.


Do take note [Quantum-correlated photons from semiconductor cavity polaritons]

VI. CONCLUSION

If for single systems with cubic nonlinearity strong nonlinearities are necessary for quantum blockade, then the mechanism of unconventional blockade makes it possible to achieve significant sub-Poisson statistics for particles in a dimer system. However, already for three connected systems – trimer, this effect disappears due to the large number of quantum trajectories. We have found that the introduction of cascade and complex quantum coupling into a system of three MPs makes it possible to achieve polariton blockade, both for individual MPs and for a combination of two pillars when they are correlated. Cross-correlations between two MPs arise in the presence of a third MP, which is pumped. In the absence of such a coherent mediator, even with a complex non-reciprocal connection, such correlations do not arise. The statistics on the second pillar will remain around coherent.

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VII. APPENDIX

A. Master equation in Fock states

From 2 and 3, we obtained system linear differential equations for matrix elements of the density matrix:

$$
\frac{d}{dt} \rho_{n,m',l',n,m,l} = -i\Delta_1 (m' - m) \rho_{n,m',l',n,m,l} - i\Delta_2 \rho_{n,m',l',n,m,l} - i\Delta_3 (l' - l) \rho_{n,m',l',n,m,l} - ig_{12} \sqrt{n'} (m' + 1) \rho_{n'-1,m'+1,l',n,m,l} + ig_{12} \sqrt{n} (n + 1) \rho_{n,m',l',n+1,m,l} - ig_{21} \sqrt{m'} (m' + 1) \rho_{n+1,m'-1,l',n,m,l} + ig_{21} \sqrt{m} (n + 1) \rho_{n,m',l',n+1,m,l} - ig_{31} \sqrt{l'} (l' + 1) \rho_{n+1,m',l'-1,n,m,l} + ig_{31} \sqrt{l} (n + 1) \rho_{n,m',l'-1,n+1,m,l} + ig_{32} \sqrt{m} (l' + 1) \rho_{n,m',l',n,m+1,l} + ig_{32} \sqrt{l} (n + 1) \rho_{n,m',l',n+1,m,l}
$$

$$
\rho_{n,m',l',n,m,l} - iU_1 (n' (n' - 1) - n (n - 1)) \rho_{n,m',l',n,m,l} - iU_2 (m' (m' - 1) - m (m - 1)) \rho_{n,m',l',n,m,l} - iU_3 (l' (l' - 1) - l (l - 1)) \rho_{n,m',l',n,m,l}
$$

$$
+ F_2 \sqrt{m'} \rho_{n,m',l'-1,n,m,l} - \sqrt{m} \rho_{n,m',l',n,m,l} + 2\gamma_1 (n' + 1) \rho_{n+1,m',l',n+1,m,l} - \gamma_1 (n + 1) \rho_{n,m',l',n+1,m,l} + 2\gamma_2 \sqrt{m} \rho_{n,m',l',n+1,m,l} - \gamma_2 (n + 1) \rho_{n,m',l',n+1,m,l} + 2\gamma_3 \sqrt{l'} \rho_{n,m',l'+1,n,m,l} - \gamma_3 (l' + 1) \rho_{n,m',l',n,m,l}
$$

$$
(7)
$$

B. Derive of the master equation for cascaded coupled

The quantum Langevin equations for cascade system are show in Fig. ?? has the follow form [35]:

$$
\frac{da_1}{dt} = -i_\hbar [a_1, H] - (\frac{\gamma_1 + \gamma_2}{2}) a_1 + \sqrt{\kappa_1} b_{in}
$$

$$
\frac{da_2}{dt} = -i_\hbar [a_2, H] - (\frac{\gamma_1 + \gamma_2}{2}) a_2 + \sqrt{\kappa_1} a_1 + \sqrt{\kappa_2} b_{in}
$$

$$
\frac{db_{in}}{dt} = F_{in} dt + dB_{in}
$$

where $b_{in} dt = F_{in} dt + dB_{in}$ is input field. Here $dB_{in}$ is quantum noise operator. Also $\gamma_1 = b_{out}^{(1)}$ and $\gamma_2 = b_{out}^{(1)}$.

From Eq. [? ] and [? ] we can turn to Ito stochastic differential equations (ISDE):

$$
da_1 = -i_\hbar [a_1, H] dt - (\frac{\gamma_1 + \gamma_2}{2}) a_1 + \sqrt{\kappa_1} F_{in} dt - \sqrt{\kappa_1} dB(t) - \sqrt{\gamma_1} dW(t)
$$

$$
da_2 = -i_\hbar [a_2, H] dt - (\frac{\gamma_1 + \gamma_2}{2}) a_2 + \sqrt{\kappa_1} a_1 + \sqrt{\kappa_2} a_1 dt - \sqrt{\gamma_2} dW(t)
$$

(10)
We can rewrite Eqs. 10 and 11 in general view as:

\[ da = -\frac{i}{\hbar} [a, H]dt - [a, a_1]\left(\frac{\gamma_1 + \gamma_2}{2} a_1 + \sqrt{\kappa_1 F_{in}}\right)dt - \\
- [a, a_2]\left(\frac{\gamma_1 + \gamma_2}{2} a_2 + \sqrt{\kappa_2 F_{in}}\right)dt - \\
-\frac{1}{\sqrt{2}} [a, \sqrt{\kappa_1} a_1 + \sqrt{\kappa_2} a_2] dB(t) - \\
-\frac{1}{\sqrt{2}} [a, \sqrt{\kappa_1} a_2 + \sqrt{\kappa_2} a_1] dB(t) \tag{12} \]

based on the follow relation \( \langle \frac{da}{dt} \rangle = \langle \frac{dp}{dt} \rangle \) and ISDE \([?]\), we can obtained the cascade master equation:

\[ \frac{d\rho}{dt} = \frac{i}{\hbar} [\rho, H] + \sqrt{\frac{\kappa_1}{2}} D(a_1) + \sqrt{\frac{\kappa_2}{2}} D(a_2) - \\
-\sqrt{\kappa_1 \kappa_2} [a_1, a_2] + \sqrt{\kappa_1 \kappa_2} [a_2, a_1] - \\
-\frac{1}{2} [a_1^2 a_2 - a_2^2 a_1, \rho] \tag{13} \]

and properties of cyclic permutations in the trace when it is resolved. The trace of the operator function 14 is equal to zero. Therefore, in a system with a non-Hermitian non-reciprocal interaction \( a_1^2 a_2 - a_2^2 a_1 \) and a non-local dissipative operator \( D(a_1 + a_2) \), the trace is preserved.

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