Biexciton-mediated superradiant photon blockade

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The photon blockade is a hallmark of quantum light transport through a single two-level system that can accommodate only one photon. Here, we theoretically show that two-photon transmission can be suppressed even for a seemingly classical system with large number of quantum dots in a cavity when the biexciton nonlinearity is taken into account. We reveal the nonmonotonous dependence of the second-order correlation function of the transmitted photons on the biexciton binding energy. The blockade is realized by proper tuning the biexciton resonance that controls the collective superradiant modes.

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Introduction.—A two-level system can absorb or emit only one photon at a time [1, 2]. As such, it is an ultimate quantum playground for the photon blockade realization [3]. Antibunching of transmitted photons in the photon blockade regime has been recently demonstrated for a cavity strongly coupled to a single atom [4–6] and to a single quantum dot [7–11]. Increasing the number of dots, one brings the system towards the classical limit. In particular, after the first photon has been absorbed, the second one can be still accommodated in one of the unoccupied dots and the photon blockade is destroyed. Indeed, the previous theoretical results for atoms without a cavity yield the absence of photon blockade [12, 13]. While the photon bunching for ensemble of two-level atoms strongly coupled to a cavity has been predicted in Ref. [14], the fundamental question of photon blockade feasibility for multiple quantum emitters remains open. Existing studies of photon blockade in cavities were limited to the case of single resonant atom [15, 16] or in a waveguide [25–27]. However, their straightforward application to multiple QDs with biexciton nonlinearity in a cavity appears to be complicated. The intuitive representation of the two-photon scattering process given by Feynman diagram technique [28] allows one to obtain the scattering amplitude only in the lowest orders of perturbation theory. Here, in order to exactly sum the perturbation series, we regard the operators \( b_{i, \sigma} \) as bosonic. Correspondence with the problem where the operators \( b_{i, \sigma} \) describe two-level systems is established by adding to the Hamiltonian an auxiliary interaction term.

Method.—Elaborate theoretical methods including Bethe ansatz [12, 13, 19], Lippman-Schwinger equation [20], input-output formalism [21], Green’s functions [22], and path integral [23, 24] have been previously used to study two-photon transport in quantum systems, such as atoms in a cavity [15, 16] or in a waveguide [25–27]. However, their straightforward application to multiple QDs with biexciton nonlinearity in a cavity appears to be complicated. The intuitive representation of the two-photon scattering process given by Feynman diagram technique [28] allows one to obtain the scattering amplitude only in the lowest orders of perturbation theory. Here, in order to exactly sum the perturbation series, we regard the operators \( b_{i, \sigma} \) as bosonic. Correspondence with the problem where the operators \( b_{i, \sigma} \) describe two-level systems is established by adding to the Hamiltonian an auxiliary interaction term.

\[ H = \sum_{\sigma=\pm} \left\{ \omega_c c_\sigma^\dagger c_\sigma + \sum_{i=1}^{N} \left[ \omega_x b_{i, \sigma}^\dagger b_{i, \sigma} + g (b_{i, \sigma}^\dagger c_\sigma + c_\sigma^\dagger b_{i, \sigma}) \right] \right\} \]

\[ - B \sum_{i=1}^{N} b_{i, \sigma}^\dagger b_{i, \sigma} + b_{i, \sigma}^\dagger b_{i, \sigma} + \right] , \quad (1) \]

where \( c_\pm \) are the annihilation operators corresponding to the two degenerate circularly polarized cavity modes with the frequency \( \omega_c \), \( b_{i, \pm} \) is the lowering operator of the two-level system corresponding to annihilation of an exciton with spin projection \( \pm 1 \) in the \( i \)-th QD, \( \omega_x \) is the exciton resonance frequency, \( g \) is the exciton-photon coupling strength, and \( B \) is the biexciton binding energy [17, 18]. The dagger denotes Hermitian conjugation and \( \hbar = 1 \).

We suppose here that all QDs are identical and neglect the polarization splitting of the cavity mode and exciton for simplicity.

\[ \text{FIG. 1. A sketch of the two-photon transmission through a cavity with } N \text{ QDs. The inset illustrates the level scheme of a QD with the biexciton nonlinearity.} \]
and leaving the cavity (crosses in Fig. 2) is equal to two-photon scattering. The dots represent interaction of two ing/exiting the cavity. (b) The series corresponding to the photon transmission. Wavy line is the bare cavity mode

FIG. 2. (a) Diagrammatic representation of the single-photon transmission. Wavy line is the bare cavity mode Green’s function Eq. (2), the crosses indicate photon entering/exiting the cavity. (b) The series corresponding to the two-photon scattering. The dots represent interaction of two excitons.

\[ \sum_{i=1}^{N} V^{\dagger}_{i,\sigma} d_{i,\sigma} + \text{and leaving the cavity (crosses in Fig. 2) is equal to two-photon } \]

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One can see that the poles of the amplitude of the second rung of the Tavis–Cummings ladder. Finally, the poles of the amplitude corresponding to two-particle excitations. For the amplitude of excitons in all QDs (superradiant mode [14, 34–38]) it indicates from interaction of the symmetric superposition of two-particle excitations. For the amplitude of excitons in all QDs (superradiant mode [14, 34–38]) (Fig. 4b), aiming at the photon blockade effect.

FIG. 3. The energies of the two-photon resonances of the scattering amplitude for a cavity with \( N = 2 \) QDs as functions of the biexciton binding energy \( B \). The solid blue (red) curves show the states relevant for excitation by two photons with parallel (opposite) spins and correspond to the poles of Eq. (7) [Eq. (8)]. The black dotted line is the energy of the bare biexciton state. The yellow dashed line shows the energy of excitation, corresponding to the single-photon resonance. The dot indicates the optimal binding energy. Right panel sketches the photon blockade effect.

at \( B = 0 \). In the limit of large binding energy \( B \to \infty \) interaction with biexciton state is suppressed so a \( \sigma^+ \) photon and a \( \sigma^- \) one interact with the system exactly in the same way as two \( \sigma^+ \) photons. Thus, we have \( M_{2\uparrow} = M_{1\uparrow} / 2 \), where the factor 2 is due to indistinguishability of photons with parallel spins.

The poles of the scattering amplitude correspond to the eigenstates of the system. We start from analysis of the one-photon resonances, being the poles of the factors \( s(\omega_a) \) in the amplitudes Eq. (7)–(8) and the transmission coefficient Eq. (3). At \( \omega_a = \omega_x \), \( \Gamma_a = \Gamma_c = 0 \) the one-particle eigenfrequencies read \( \omega_a = \omega_x \pm \sqrt{N} g \), which corresponds to the energies of the first rung of the Tavis–Cummings ladder [33]. The splitting \( 2\sqrt{N} g \) originates from interaction of the symmetric superposition of excitons in all QDs (superradiant mode [14, 34–38]) with a cavity photon. In addition to these one-particle resonances, the scattering amplitude has also poles corresponding to two-particle excitations. For the amplitude \( M_{1\uparrow \downarrow} \) they read \( 2\epsilon = 2\omega_x \pm 2g \sqrt{N} - 1/2 \), being the energies of the second rung of the Tavis–Cummings ladder. Finally, the poles of the amplitude \( M_{1\uparrow \uparrow} \), and the next poles of \( |\psi_{\text{out}}(\omega)\rangle = |\psi_{\text{out}}^{(1)}(\omega)\rangle\rangle_{\alpha a} |\psi_{\text{out}}^{(2)}(\omega)\rangle\rangle_{\alpha a} \), where \( a_x(t) \), \( \omega_x \rangle \), \( \langle \omega_x \rangle \) correspond to the poles of the one-particle resonance and maximizes one-photon transmission. This is in contrast to off-resonant excitation with the frequency \( \omega_x \), considered in Ref. [14]. In our case

\[
\begin{align*}
g^{(2)}(0) &= \frac{2N^{3/2}g - (2N + 1)B + 4iN^2g}{4iN^3g - \sqrt{N}B(g(\Gamma + 2\sqrt{N})} \left| \frac{1}{\sqrt{N}B(g(\Gamma + 2\sqrt{N})} \right|^2, \\
\end{align*}
\]  

where \( \Gamma = \Gamma_c + \Gamma_x \). Expression Eq. (10) for the two-photon correlations is our main result. Next, we analyze its dependence on the number of dots \( N \) (Fig. 4a) and the biexciton binding energy \( B \) (Fig. 4b), aiming at the photon blockade condition \( g^{(2)}(0) < 1 \).

The thick blue curve in Fig. 4a shows the behavior of \( g^{(2)}(0) \) with \( N \) for \( B \to \infty \) corresponding to absence of biexciton resonance. At \( N = 1 \) one has \( g^{(2)}(0) \approx 1 \), reflecting the photon blockade. The essence of the photon blockade effect is the detuning of the two-photon transmission resonance from double energy of single-photon
resonance [4]. Interestingly, the blockade is conserved even for multiple QDs in the cavity and destroyed only in the classical limit \( N \to \infty \). This is in contrast to the case of QDs without a cavity where the blockade vanishes at \( N \geq 2 \), cf. thick blue and thin black curves in Fig. 4a. The photon pair in cavity could excite only the symmetric superradiant state of the Tavis–Cummings ladder \( 2\omega_x - 2\sqrt{N - 1/2} g \) (red curve in Fig. 3) that is however detuned from the double energy of excitation \( 2\varepsilon = 2(\omega_x - \sqrt{Ng}) \) (yellow dashed line). The photon blockade persists up to relatively large \( N \sim (g/2\Gamma)^2 \sim 25 \) when the detuning becomes less than the level width \( \Gamma \).

The effect of biexciton binding energy \( B \) on the photon blockade is shown in Fig. 4b. Surprisingly, the dependence of the second order correlation function on the biexciton energy is nonmonotonous: for certain binding energy \( B^* \) the minimal value of \( g^{(2)} \) is achieved. Moreover, the dip in the dependence does not disappear with increase of \( N \), meaning that the antibunching can be achieved even for large numbers of QDs. The reason is the biexciton-induced detuning of the two-particle superradiant state. Namely, for \( N \ll (g/2\Gamma)^2 \) the photon blockade is enhanced due to the interference of the transmission channels of photons with parallel and opposite spins, \( M_{xx,xx} = M_{\uparrow \uparrow} + \frac{1}{2} M_{\uparrow \downarrow} \). The maximal antibunching \( g^{(2)}(0) = 4(2N + 1)^2(\Gamma/g)^4 \) is achieved at the optimal binding energy \( B^* = N^{3/2}g/(N + 1/2) \), see yellow dot in Fig. 3. This minimal value of \( g^{(2)}(0) \) is proportional to \( (\Gamma/g)^4 \) and parametrically smaller than that for the absence of biexciton resonance (at \( B = \infty \) one has \( g^{(2)}(0) = (2N + 1)^2\Gamma^2/Ng^2 \), cf. solid red and blue curves in Fig. 4a. For large \( N \gg (g/2\Gamma)^2 \) the correlation function \( g^{(2)}(0) \) is equal to unity for all biexciton binding energies except for the narrow region of the width \( \sim g \) around \( B^* = 2\sqrt{Ng} \) corresponding to the anticrossing of the biexciton level with the second rung of the Tavis–Cummings ladder. In this case the double excitation energy hits exactly into the energy gap between the split \( \uparrow \uparrow \) superradiant modes, see Fig. 3. The splitting \( \sim g \) being larger than the level width \( \sim \Gamma \), the photon blockade is recovered in the \( \uparrow \downarrow \) channel, i.e. transmission of two counter-polarized photons is suppressed. The \( \uparrow \uparrow \) channel is not coupled to the biexciton resonance, and does not manifest the photon blockade. The total value of \( g^{(2)}(0) \) for linearly polarized photons, contributed by both \( \uparrow \uparrow \) and \( \uparrow \downarrow \) channels, is equal to 1/4, see the red curve in Fig. 4a.

To summarize, we have demonstrated that the photon blockade regime is realized for the polarization-dependent two-photon transport through a seemingly classical system of a microcavity with multiple quantum dots. Two-photon correlation function of transmitted photons is a nonmonotonous function of the biexciton resonance energy. Even for a cavity with many quantum dots the proper tuning of biexciton energy allows one to achieve the values of \( g^{(2)}(0) \) that are as low as 1/4. Experimental realization of the blockade requires the inhomogeneous broadening of exciton frequencies to be small enough so that the collective superradiant exciton mode exists. This is feasible because the simultaneous strong coupling regime of the cavity mode with up to three quantum dots has been already reported [39–42].

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FIG. 4. (a) Dependence of photon correlation function \( g^{(2)}(0) \) on the number of QDs \( N \). Thin black curve corresponds to reflection from QDs without cavity (or, alternatively, a cavity with QDs in a weak coupling regime). Thick blue and red curves show the correlation function of \( x \)-polarized transmitted photons for a cavity with \( N \) QDs in the strong coupling regime, \( \Gamma = 0.1g \), under \( x \)-polarized excitation with the frequency \( \varepsilon = \omega_x - \sqrt{Ng} \). Thick blue curve corresponds to absence of biexciton state \( (B = \infty) \), while the thick red one corresponds to biexciton being tuned to the optimal energy, \( B = B^* \), that maximizes the photon blockade. (b) Dependence of \( g^{(2)}(0) \) on the biexciton binding energy \( B \) for cavity with different numbers of QDs in the strong coupling regime, \( \Gamma = 0.1g \).
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