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Abstract. A scheme to observe direct experimental evidence of Jaynes–Cummings nonlinearities in a strongly dissipative cavity quantum electrodynamics system was devised. In such a system, large losses compete with the strong light-matter interaction. Comparing coherent and incoherent excitations of the system, it was shown that resonant excitation of the detuned emitter makes it possible to evidence few photon quantum nonlinearities in currently available experimental systems. [DOI: 10.1117/1.JNP.6.061803]

Keywords: Jaynes-Cummings; microcavity; photon statistics; polariton; quantum dot.

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1 Introduction

The physics of light-matter coupling in semiconductor heterostructures[1] finds its vantage point on quantum physics in the zero-dimensional case.[2] After reaching the strong coupling regime for optically active quantum dots (QDs) in high quality microcavities[3–5] and demonstrating its single-photon character,[6,7] a major remaining challenge is to obtain clear and direct evidence of quantum nonlinearities. Tailoring the interaction of light with matter at the single-quantum level is one of the most pressing goals for both fundamental and applied research. A successful implementation sees a single quantum of excitation affecting the response of the system. Indirect manifestations have already been provided in the form of photon blockade[8–10] or broadening of the Rabi doublet due to excited states,[11] but these are unspecific with regard to their origin and whether such quantum effects are described by the Jaynes–Cummings (JC) Hamiltonian,

$$H = \omega_a a^\dagger a + (\omega_a - \Delta)\sigma^\dagger \sigma + g(a^\dagger \sigma + a\sigma^\dagger), \tag{1}$$

the paradigm of quantum interaction between quanta of light (with Bose operator a) and a two-level system (σ). Such quantum nonlinearities have been demonstrated in cavity QED systems, such as atoms[12,13] or, perhaps most spectacularly, for superconducting qubits,[14,15] where the fingerprints of JC physics have been observed, in particular the anharmonic splitting of states with the number of excitations.[16] Only very recently have semiconductor systems begun to exhibit this rich phenomenology.[17,18] The difficulty of directly observing transitions between the different rungs of the JC ladder in semiconductors can, presumably, be traced to the strong dephasing in these systems. Indeed, in the spectral domain, the uncertainty due to the short photon lifetime washes out completely the weak square-root dependence of the splitting of the excited states. Any traces of the quantum interaction, lost in the energy of the emitted photons, is however recovered in the statistics of the emitted photons.[19]

We will highlight that, although it is difficult to obtain clear evidence of the JC ladder in state-of-the-art semiconductor samples when detecting luminescence under incoherent excitation, it is possible to observe clear signatures of the higher rungs by performing photon-counting measurements to probe the statistics of the emitted photons from the cavity. We show that this

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process is optimum when coherently exciting the detuned QD, which results in strong photon bunching at the resonances of the JC ladder. Our results provide a route for experimentalists to test the suitability of JC model to describe QD-cavity systems, which have displayed many variations from their counterparts in atomic or superconducting cavity QED.[20,21] The scheme can also be used as a source of quantum light, generating hugely bunched statistics.

2 Dissipation in Quantum Mechanics

The effect of dissipation can be introduced into the JC Hamiltonian with Liouville equation $\partial_t\rho = \mathcal{L}(\rho)$, where the so-called Liouvillian $\mathcal{L}$ adds a nonunitary evolution of the density matrix ρ to the Hamiltonian dynamics:

$$\mathcal{L}(\rho) = i[\rho, H] + \frac{\gamma_a}{2}\mathcal{L}_a(\rho) + \frac{\gamma_\sigma}{2}\mathcal{L}_\sigma(\rho) + \frac{P_a}{2}\mathcal{L}_{a^\dagger}(\rho) + \frac{P_\sigma}{2}\mathcal{L}_{\sigma^\dagger}(\rho), \tag{2}$$

where $\mathcal{L}_c(\rho) = 2c\rho c^\dagger - c^\dagger c\rho - \rho c^\dagger c$. This describes the decay (at rate γ_a for the cavity photon and γ_σ for the QD exciton) or excitation (P_a and P_σ) due to incoherent pumping.[22] Dephasing can be included in this formalism with additional terms $\mathcal{L}_{\sigma^\dagger\sigma}$ for pure dephasing[5,23,24] or $\mathcal{L}_{\sigma^\dagger a}$ for phonon-induced dephasing.[25] We have checked that unless these quantities have very large values, they do not qualitatively affect our findings.

As a result of finite lifetime, the energies of the JC system become complex. They are obtained by diagonalizing the Liouvillian Eq. (2):

$$E_\pm^k = k\omega_a - \frac{\Delta}{2} - i\frac{(2k-1)\gamma_a + \gamma_\sigma}{4} \pm \sqrt{\left(\sqrt{k}g\right)^2 + \left(\frac{\Delta}{2} - i\frac{\gamma_a - \gamma_\sigma}{4}\right)^2}, \tag{3}$$

where $E_\pm^k$ corresponds to the kth rung of the system. This is a generalization of the usual expression that neglects lifetime. Following Eq. (3), we plot the eigenenergies of a dissipative JC system (for $\gamma_a = g$ and $\gamma_\sigma = 0$) in Fig. 1 as a function of detuning. Here, the broadening $\pm 2\Im(E_\pm)$ is visualized as the width of the line. Note that the spacing between two consecutive rungs has been shrunk for comfort of visualization, and is in reality two or three orders of magnitude larger than shown (the case where the energy between two manifolds becomes comparable with the polariton splitting exists but belongs to an altogether different regime, called "ultrastrong coupling").[26,27] The first rung ($k = 1$) is the familiar anticrossing of two coupled modes, that describes equally well the linear quantum regime and a classical system.[28] Higher rungs ($k > 1$) reproduce the same pattern with two variations with respect to the linear case (at resonance): the splitting increases as $\sqrt{k}$ and the broadening as $k\gamma_a/g$. Because the increase of the coupling rate with the number of excitations (k) is slower than the decoherence, climbing the ladder makes it increasingly difficult to observe the quantum features. One could circumvent this problem by decreasing γ_a, but in typical semiconductor systems $\gamma_a \approx g \gg \gamma_\sigma$. However, it is advantageous if γ_a is not too small: it increases signal intensity and better preserves the statistics of the state prepared inside the cavity. These two qualities are essential for working quantum devices.

Equation (3) shows how the coupling, decay, and detuning coexist in the polariton. The real part of $E_\pm^k$ yields its energy whereas the imaginary part yields its (inverse) lifetime. The part outside of the square root is clear and intuitive. There is, however, a rich physics from the quantum coupling in the complex-valued square root. Detuning alone further splits the polaritons (because the bare states are themselves brought apart), whereas decay alone decreases the splitting and eventually makes the square root imaginary, mark of the weak coupling. When both detuning and decay appear together, they contribute a joint imaginary term from the double product, which washes out the notion of weak- and strong-coupling, some aspects of which are presented in Fig. 2. The real part (a) and the imaginary part (b) of the splitting in the various manifolds between polaritons in the kth rung are plotted for $\gamma_\sigma = 2g$, which is where the maximum splitting (of the dressed states) is observed as a function of γ_a. At resonance, there is a clear criterion to define strong-coupling in the kth rung, namely, when the states are split in energy and have the same linewidth. The transition to weak-coupling results in a sharp nondifferentiable jump between an analytic function and zero. With detuning, however, this sharp transition smoothes out and the curve becomes differentiable

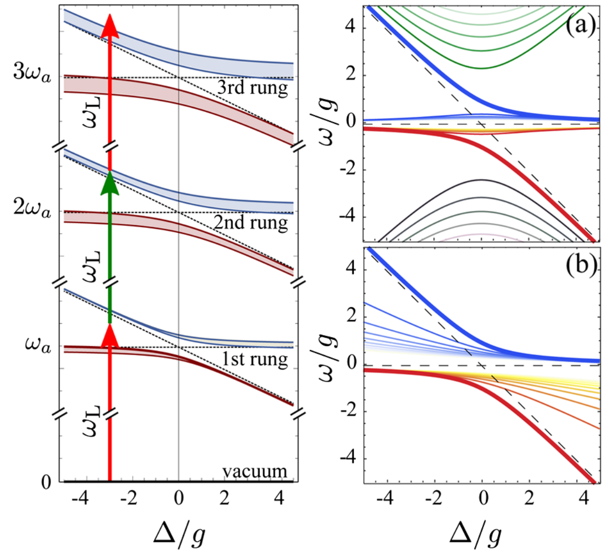

Fig. 1 Left: Energies and linewiths of the first three rungs of the dissipative JC ladder (for $\gamma_a = g$ and $\gamma_\sigma = 0$) as a function of detuning. The configuration of the two-photon blockade is indicated by the arrows. Right: Transition energies and resonances of the JC ladder as a function of detuning, probed under incoherent (a) and coherent (b) excitation, respectively. The thick solid lines are the upper and lower polariton lines of the first rung. The thin dotted lines are the bare (undressed) QD and cavity. They sandwich inner lines and are encompassed by outer lines from transitions higher in the ladder.

everywhere, recovering the bare states only asymptotically rather than, previously, exactly. The difference between the splitting of the detuned light-matter coupled states and the bare states in Fig. 2(c) shows that the coupled states with dissipation recover the bare state only in the case of resonance or infinite Δ. With detuning, even in weak coupling, the states are thus more splitted than the bare states.

3 Observables

Experimentally, the energy structure specified by Eq. (3) cannot be observed directly, and the way it manifests itself depends on the kind of measurement performed and how the system is

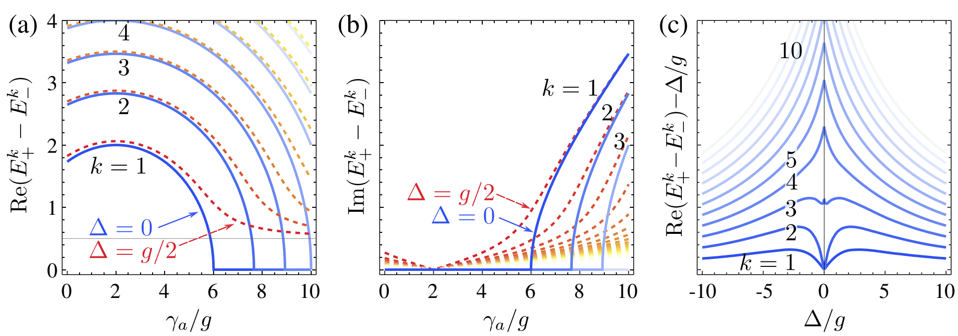

Fig. 2 Behavior of the real part (energy splitting) (a) and imaginary part (difference of broadening) (b) of the Rabi splitting between polaritons in the kth manifold, with $\gamma_\sigma = 2g$. At resonance (solid blue), strong-coupling in the kth rung is well defined by splitting of the states with identical broadening. Detuning (dashed red) mixes weak- and strong-coupling features at all coupling strengths. In (c), the difference between the kth manifold Rabi splitting is compared to that of the bare states for $\gamma_a = 8.55g$. The first two rungs are in weak-coupling, resulting in the two lower curves being exactly zero at $\Delta = 0$. With detuning, however, the states experience the cross detuning-decay term and start to split away from the bare states. This also has incidence on the third rung, by perturbing the splitting around resonance, first decreasing, then increasing it, before recovering the normal trend of higher rungs.

excited. There are countless variations of experiments, but most can be categorized in incoherent and coherent excitation. In the former case, one populates the energy levels of the system through relaxation of charge carriers and observes the emission at energies corresponding to transitions between consecutive manifolds. In the latter case, a well-defined energy is incident on the system, and one observes its direct response via a number of observables. The expected resonances of the system in these two configurations are displayed in Fig. 1(a) and 1(b), respectively. Both have in common the upper polariton (UP) and lower polariton (LP) lines (thick lines) of the first rung, which are most easily excited and detected. To prove the quantum character of this system, the field quantization needs to be demonstrated, and this requires observing at least some of the lines that arise from higher rungs of the ladder.

3.1 *Incoherent Excitation*

In the incoherent excitation case [Fig. 1(a)], there are two sets of additional lines: one in between the Rabi doublet, the other sandwiching it. The inner lines are narrowly packed together and are broader, since they arise from higher rungs, thus demanding an extraordinarily good system to resolve them. The outer lines have a much higher splitting between them. However, the strength of these transitions is strongly suppressed in the cavity emission, since the emitter is de-excited at the same time as the photon is emitted, while the cavity favors the sole emission of a photon. To climb the ladder in a dissipative system, a rather strong excitation is required, and this can strongly renormalize the energy levels, broaden them, or on the opposite, narrow them as a result of Bose-stimulation and onset of lasing. In Fig. 3, we show the cavity photoluminescence spectra $\langle a^\dagger(\omega)a(\omega)\rangle$ (Ref. 29) as function of detuning for increasing dissipation from top to bottom and for various intensities of incoherent excitation from left to right. Axes are not shown for clarity but are the same as Fig. 1(a). The quality of the strong-coupling ranges from significantly better than is currently available ($\gamma_a/g = 0.1$), via state-of-the-art systems [$\gamma_a/g = 0.5$ (Ref. 30)], to the typical value available in many laboratories worldwide ($\gamma_a/g = 1$ [Ref. 23]). These density plots show how the JC structure of Fig. 1(a) manifests itself in photoluminescence. In all cases, the outer lines are indeed suppressed. The main features are the upper and lower polaritons.

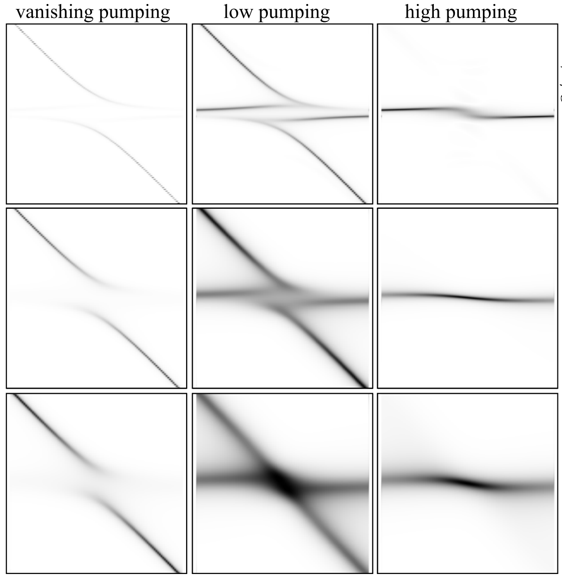

Fig. 3 Incoherent excitation: cavity photoluminescence spectra for increasingly dissipative systems from upper to lower row ($\gamma_a/g = 0.1$, 0.5 and 1), and for increasing excitation power from left to right column. Quantum nonlinearities are more clearly observed for a small, but nonvanishing, incoherent excitation. Higher pumping brings the system into lasing or collapses the Rabi doublet. Only in very strongly coupled systems does the photoluminescence reconstruct the JC ladder, albeit with the outer transitions being much suppressed in the cavity emission. Axes are not shown for clarity but are the same as Fig. 1(a).

When one tries to climb the ladder by increasing pumping, only in the very best system can additional lines of the second rung be clearly resolved. In the case of $\gamma_a/g = 0.5$, although a strong deviation from the anticrossing is observed, no clear fingerprints of the JC features are observed. At resonance, only a doublet is observed (qualitatively similar to the Rabi doublet), and out of resonance, a triplet is observed.[31] This might in fact be consistent with the experimental situation of Ref. 20, that did not make any claim in this direction. For smaller strong-coupling, although still deviating from crossing or anticrossing, there is again no useful characterization of the JC physics. At pumping levels higher than those presented in Fig. 3, the system moves into the lasing regime[30] and the JC description is not adequate anymore.[32]

3.2 *Coherent Excitation*

In the coherent excitation case [Fig. 1(b)], there are only inner lines, but they are clearly separated from one another in the energy-detuning space. These arise from multiple-photon excitations. When the laser is at an energy ω_L, smaller than the upper polariton ω_{UP} energy, as indicated by the arrows in Fig. 1, it cannot excite the system with one photon. However, if $2\omega_L = \Re(E_+^2)$, then it can access the second rung by a two-photon excitation process. The separation of the lines is larger at larger detunings, and the broadening of the exciton-like polariton is also smaller than at resonance, where it is half cavity-photon. However, the coupling is maximum at resonance. There is a tradeoff for the detuning between a small dephasing of the states and a good coupling. We will show that the advantages of detuning predominate: if nonlinearities can be evidenced, the loss of maximum admixtures of light and matter is not detrimental.

At the point highlighted, the laser is blocked at the first rung, is resonant with the second rung, and is blocked again at the third rung, even when taking into account the large broadening of higher excited manifolds. This configuration, therefore, efficiently filters out the two-photon fluctuation of the laser and performs a type of two-photon blockade, in analogy with the photon blockade effect,[33,34] where blocking from the second rung is used to produce a single-photon source.[8,9] This scheme does not work so well at resonance because of overlap of the transitions broadened by dissipation.

Furthermore, as for incoherent excitation, one should try to avoid overly strong pumping for coherent excitation. In the Hamiltonian, it is included by adding the term $\Omega_a \exp(i\omega_L t)a^\dagger + \mathrm{h.c.}$ for coherent cavity excitation.[8–10] One can also excite the QD coherently $\Omega_\sigma \exp(i\omega_L t)\sigma^\dagger + \mathrm{h.c.}$, e.g., by side emission in a pillar microcavity. At low driving intensity, one only sees clearly the lower and upper polariton of the first rung, but for increasing pumping the coherent excitation quickly dresses the states and distorts Eq. (3). In the first column of Fig. 4 we plot the cavity population $n_a = \langle a^\dagger a\rangle$ when driving the system coherently for increasingly dissipative systems from top to bottom. The plots fail to reproduce the nonlinear features of Fig. 1(b), indicating that the intensity ($\propto n_a$)—and in fact other observables involving first-order correlators such as reflectivity, transmission, absorption, etc.—is not optimal to resolve the higher rungs of the JC ladder. In all these cases, there is a clear observation when the laser hits the polariton resonances of the first rung, but otherwise the response is weak. There is, however, a strong response in observables involving higher-order correlators (at zero time delay) of the type $G^{(n)} = \langle a^{\dagger n} a^n\rangle$, which are linked to photon counting. One usually deals with the normalized quantities, $g^{(n)} = G^{(n)}/n_a^n$. In particular, $g^{(2)}$ is popular as the standard to classify antibunched (nonclassical) ($g^{(2)} = 0$), Poissonian (coherent) ($g^{(2)} = 1$), and bunched (chaotic/thermal) ($g^{(2)} = 2$) light sources. Other related quantities are more useful in this context,[35] such as the Mandel parameter Q_{M}:

$$Q_M = (g^{(2)} - 1)n_a, \tag{4}$$

which sign provides the (anti)bunched character of the statistics, while also taking into account the available signal. The Q_M parameter of the light emitted by the cavity is shown in Fig. 4 for coherent excitation of the QD (middle column) and of the cavity (right column). Remarkably, the middle column shows that this measurement unravels the second rung of the JC ladder: when the coherent excitation source is tuned to the first or the second resonance, the photon statistics sharply responds. As detailed in Fig. 5(a) and 5(b), the statistics changes its character from

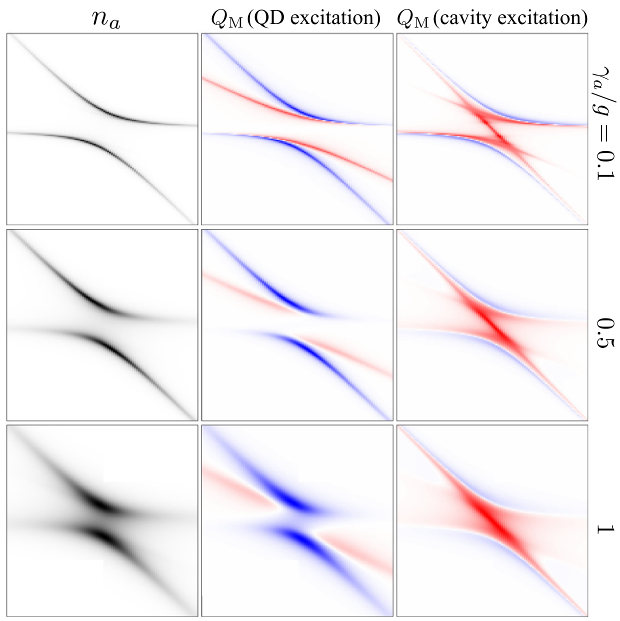

Fig. 4 Coherent excitation: cavity intensity n_a (first column) and Mandel factor Q_M under coherent QD (second column) and cavity (third column) excitation, for increasingly dissipative systems. Blue and red refer to negative (antibunching) and positive (bunching) values, respectively. Whereas intensity (or other observables such as absorption or scattering) elicits a response only from the bottom of the ladder, the photon statistics displays strong features from the second rung when exciting the QD detuned from the cavity. The signature is then unambiguous even for very dissipative systems. For cavity excitation, bare modes dominate. Axes are not shown for clarity but are the same as Fig. 1(b).

antibunching on the polariton to bunching on the second rung, caused by the two-photon blockade configuration displayed in Fig. 1. As a result of the separation of the resonances, one can clearly observe them both in this measurement with QD excitation. The scheme is much less efficient when exciting the cavity, with a strong response on the bare QD, as can be seen both on the density plot in Fig. 4 (right column) or on the cut in Fig. 5(a) and 5(b). This is due to a sudden drop of the intensity when exciting the cavity at the energy of the QD.

These results show that even in very dissipative systems, where the coupling rate is of the order of the decay rate, the second rung can be unambiguously observed in experiments by combining detuning, QD excitation, and photon counting. Although it is beneficial to maximize the coupling, there is in fact no limiting factor from strong or weak coupling in spontaneous emission. In this way, this scheme can be extended to higher rungs of the ladder, even though lower ones are in weak coupling. In this text, for the sake of brevity, we shall consider only strong-coupling situations, however.

Measuring the differential correlation function:

$$C^{(n)} = \langle a^{\dagger n} a^n \rangle - \langle a^\dagger a \rangle^n, \tag{5}$$

which quantifies the deviation of coincidences from uncorrelated, Poissonian events, one finds sharp resonances at the nth photon resonance condition. In Fig. 5(c), we plot $C^{(n)}$ up to the fourth rung, still for coherent excitation of the dot, when it is detuned by $\Delta/g = 4$ from the cavity. Such high-order coincidences can be measured by recently developed experimental techniques such as photon counting using a streak camera.[36] The signal is increasingly difficult to obtain for higher orders, as it requires the accumulation of statistics for increasingly unlikely events (curves have been rescaled as indicated in the figure). However, given a sufficiently strong signal, one obtains sharp resonances precisely located at the JC multiphoton resonances, even for very dissipative systems such as those currently available. The scheme is robust to increased pumping, which broadens the resonances but does not appreciably shift their maxima, and provides more signal. Although the experiment to perform is the same, we wish to emphasise how $C^{(n)}$ can provide a

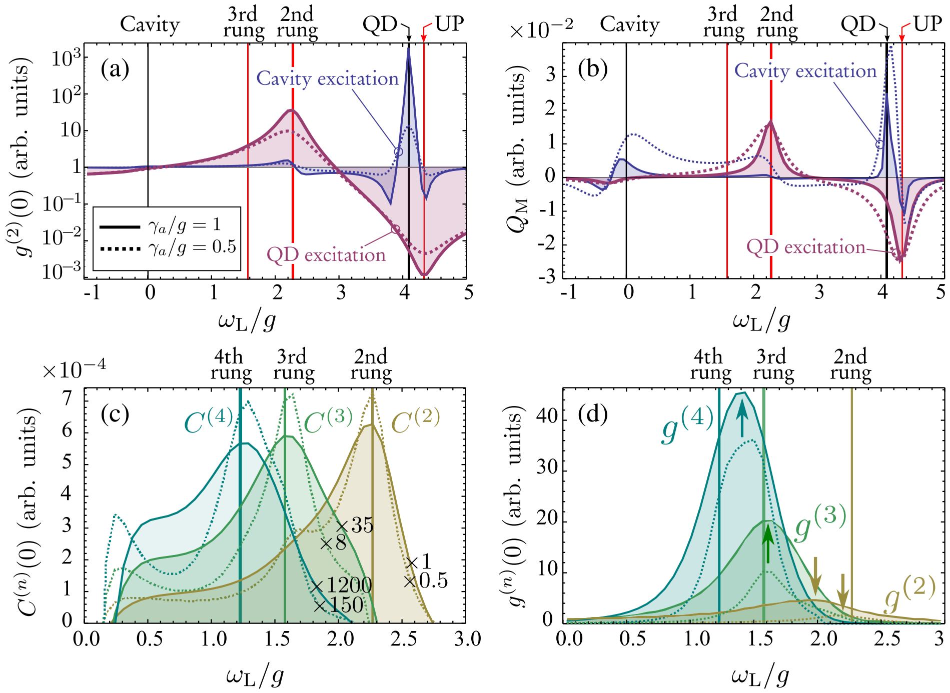

Fig. 5 Quantum statistics of the cavity photons under coherent excitation for a good (dashed lines) and a typical (solid lines) system. (a) and (b), $g^{(2)}$ and Q_M parameter, showing how the second-rung transition is more clearly identified under QD excitation. Q_M is preferable to $g^{(2)}$ as it takes into account the available signal. (c), Differential correlations $C^{(n)}$ that peak sharply at the nth resonance, thus clearly identifying the higher states of the ladder. (d), nth-order correlation functions $g^{(n)}$, that are loosely connected to the theoretical transitions. Maxima are indicated by arrows.

cleaner result than $g^{(n)}$, which is shown in Fig. 5(d) for comparison. One sees that $g^{(n)}$ increases with n, which marks the higher quantumness of light emitted when hitting the higher rungs. However, the signal also becomes exponentially dimmer as a result. Because of fluctuations to all orders, the resonances are also not exactly mapped with the transitions [compare the agreement of $g^{(2)}$ with the theoretical line at low pumping in (a) and its disagreement at high pumping in (d)], in contrast to $C^{(n)}$ that follow them more accurately (except very close to resonance).

4 Conclusion

In conclusion, we have discussed how to unravel the JC nonlinearities in dissipative QD-cavity systems with figures of merits currently available. We have shown that by analyzing the photon-counting statistics of the light emitted by a cavity when the quantum emitter is detuned and excited coherently, one can observe a clear fingerprint of the JC ladder. In view of the popularity of cavity excitation (in transmission, reflectivity, or other types of measurements), we must highlight the importance of exciting the quantum dot, which is the source of quantum nonlinearity in the coupled system, rather than the cavity mode, which is of a classical character. The engineering of JC physics of QDs in microcavities will pave the way for practical, working quantum information devices in the solid state.

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