

Second-order correlations in an exciton-polariton Rabi oscillatorYuri G. Rubo,¹ Alexandra Sheremet,^{2,3} and Alexey Kavokin^{3,4,5,6}¹*Instituto de Energías Renovables, Universidad Nacional Autónoma de México, Temixco, Morelos, 62580, Mexico*²*Information Technologies, Mechanics, and Optics University, St.-Petersburg, 199034, Russia*³*Russian Quantum Center, Novaya 100, 143025 Skolkovo, Moscow Region, Russia*⁴*School of Physics and Astronomy, University of Southampton, SO17 1BJ, Southampton, United Kingdom*⁵*Spin Optics Laboratory, St.-Petersburg State University, 198504, Peterhof, St.-Petersburg, Russia*⁶*CNR-SPIN, Viale del Politecnico 1, Rome I-00133, Italy*

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We develop the theoretical formalism to calculate second-order correlations in dissipative exciton-polariton system and we propose intensity-intensity correlation experiments to reveal the physics of exciton-light coupling in semiconductor microcavities in the Rabi oscillation regime. We predict a counterintuitive behavior of the correlator between upper and lower polariton branches: Due to the decoherence caused by stochastic exciton-photon conversions this correlator is expected to decrease below 1, while the individual second-order coherence of upper and lower polaritons exhibits nonmonotonous bunching.

DOI: [10.1103/PhysRevB.93.115315](https://doi.org/10.1103/PhysRevB.93.115315)**I. INTRODUCTION**

Recent decades have seen tremendous progress in the physics of strongly coupled light-matter systems. In particular, semiconductor microcavity structures [1] have demonstrated a number of fascinating coherent many-body effects, e.g., polariton lasing [2] and formation of Bose-Einstein condensates of exciton-polaritons [3]. Exciton-polaritons may be viewed as quantum superposition states of light and matter [4]. Their dual nature is visualised in polariton Rabi oscillations: beats between exciton and photon states in semiconductor microcavities in the strong coupling regime [5]. Usually, the Rabi oscillations are excited by a short laser pulse which simultaneously and equally populates the upper polariton (UP) and the lower polariton (LP) branches. Initially, the system is in a purely photonic state. Then, due to the splitting between UP and LP branches, it starts developing the excitonic component, becomes purely excitonic after several fractions of a picosecond, then returns to the photonic state, and so on. Polariton Rabi oscillations have been experimentally observed by many groups [6–8]. Usually, only several periods of oscillations could be resolved in these studies. The magnitude of oscillations was found to decrease with time and eventually vanish due to some decoherence processes. The nature of these processes still needs to be revealed. One process is the phonon-assisted scattering between UP and LP branches that leads to depopulation of the upper branch and breaks coherence between two branches [9]. In a recent work [10] we have addressed a process of stochastic exciton-photon conversion, which is crucial in the weak exciton-photon coupling regime and possibly plays an important role in the strong coupling regime too. The question of existence of stochastic processes even in the strong coupling regime is important for understanding the quantum properties of exciton-polaritons. To what extent polaritons can be considered as coherent superpositions of photons and excitons? How accurate would be the description of a polariton gas in terms of an exciton-photon mixture? The dynamics of exciton-photon correlators in the polariton lasing regime calculated in our previous paper [10] would help answer these questions. Unfortunately, direct experimental measurements of exciton-photon correlations

seem quite tricky. Here we show that instead of counting individual excitons and photons one can access the stochastic exciton-photon transformation kinetics by doing a purely optical and much simpler experiment. Namely, one can study two-color intensity-intensity correlations of light emitted from UP and LP branches in the strong coupling regime. The scheme of the proposed optical experiment is shown in Fig. 1.

The finite lifetime of excitons and photons does not destroy the coherence of Rabi oscillations; only the amplitude of the signal decays in time after pulsed excitation of the system. The stochastic conversion of particles, however, has a profound destructive effect on the Rabi process. The result is qualitatively similar to the suppression of tunneling for a particle in a double-well potential in the presence of dissipation [11]. The system we study below also has some similarity to the phonon-polaron problem [12–14]. We note, however, that the stochastic processes that are the focus of our present study have not been analyzed in the context of the electron two-level systems and other works on polaron-transformed master equations known to us. We note also that the bosonic two-level systems provide benefits related to the possibility of weak measurement of the system state. In our case it is achieved by study of correlations of photons emitted from the microcavity. In this paper, we present the exact theory of second-order correlations on exciton-polariton system in the presence of dissipation due to exciton-photon conversion. We predict a counterintuitive variation of the intensity-intensity correlator between UP and LP branches (two-color correlation experiment) and show that this variation may be considered as a signature of stochastic conversions in the strong coupling regime. The microscopic origin of exciton-photon conversion processes is not specified here.

We would like to point out at this point that we account for only two exciton-polariton states in the model: one belonging to the upper polariton branch and one belonging to the lower polariton branch. There is no exciton reservoir in the model, as in Rabi-oscillation experiments with resonant optical excitation the reservoir remains empty. The expected role of incoherent exciton reservoir is twofold. On the one hand, the continuous feed of the Rabi oscillator from the reservoir

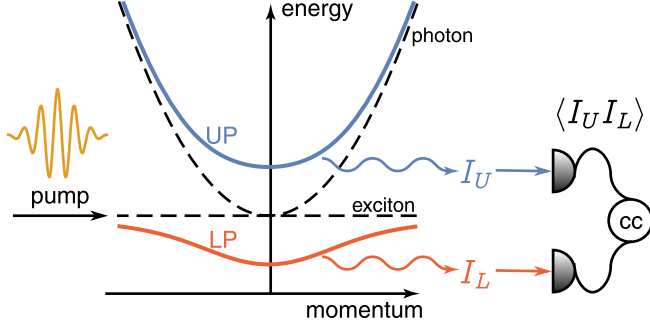


FIG. 1. The scheme of the two-color intensity-intensity correlation measurement between upper and lower polariton branches. A short pulse of light excites the system and induces polariton Rabi oscillations. Correlations of the intensities of secondary emission harmonics corresponding to upper and lower polariton branches are studied.

increases the duration of Rabi oscillation [15] and therefore facilitates the observation of even rare events of exciton-photon conversion. On the other hand, the fluctuations produced by the exciton reservoir on the Rabi oscillator are not completely described by our model [formulated in Eq. (1) below] and they will be a subject of subsequent research.

From the point of view of quantum statistics, the loss of coherence in a light source is manifested by the variation of the intensity-intensity correlator $g^{(2)}$ that ranges between 1 (purely coherent light) and 2 (thermal distribution of photons) for classical sources [16]. In the regime of polariton Rabi oscillations, the quantum coherence between two branches can be characterized by a similar quantity g_{ul} defining the correlator of intensities of light emitted from the UP and LP branches. A coherent optical excitation sets $g_{ul} = 1$ initially and simple depopulation of the branches does not affect this value. However, as we show below, the stochastic exciton-photon conversion processes manifest themselves in deviation of the UP-LP intensity correlator from unity, so that it becomes lower than 1 at large times.

II. THEORETICAL MODEL

The dissipative exciton-polariton system can be described by Lindblad form of the Liouville equation for the full density matrix [10]

$$\frac{d\hat{\rho}(t)}{dt} = \frac{i}{\hbar}[\hat{\rho}, \hat{H}_0] - \sum_j \frac{g_j}{2}([\hat{A}_j^\dagger, \hat{A}_j \hat{\rho}] + [\hat{\rho} \hat{A}_j^\dagger, \hat{A}_j]). \quad (1)$$

Here the Hamiltonian is

$$\hat{H}_0 = \frac{\hbar}{2}\{\Delta(\hat{b}^\dagger \hat{b} - \hat{a}^\dagger \hat{a}) + \omega_R(\hat{a}^\dagger \hat{b} + \hat{b}^\dagger \hat{a})\}, \quad (2)$$

where \hat{a} and \hat{b} are the exciton and photon annihilation operators, respectively, Δ is the exciton-photon detuning, and ω_R is the exciton-photon coupling frequency. In Eq. (1) we introduced three ($j = x, c, u$) single-particle Lindblad terms with $\hat{A}_x = \hat{a}$, $\hat{A}_c = \hat{b}$, and $\hat{A}_u = (\hat{a} + \hat{b})$. The first two terms describe the direct depopulation of the exciton and photon states. The third term with coefficient $g_u \equiv \gamma'$ simply renormalizes the exciton and the cavity photon lifetimes, $\tau_x = (g_x + \gamma')^{-1}$ and $\tau_c = (g_c + \gamma')^{-1}$, on the one hand. On the other hand, it

models the cross relaxation and allows phenomenologically for an additional decay from the upper polariton state that is crucial for description of realistic microcavities. Finally, there are two ($j = xc, cx$) terms describing the conversion processes, with $\tau_{xc} = \tau_{cx} = g_{xc}^{-1}$ being the exciton-photon conversion time and operators $\hat{A}_{xc} = \hat{b}^\dagger \hat{a}$ and $\hat{A}_{cx} = \hat{a}^\dagger \hat{b}$.

In what follows we shall consider different first- and second-order correlations. For the density matrix satisfying (1) it is possible to find the closed systems of equations for the correlators of any order. To proceed, it is convenient to introduce the spin operators \hat{s}_μ with $\mu = 0, 1, 2, 3$:

$$\hat{s}_0 = \frac{1}{2}(\hat{a}^\dagger \hat{a} + \hat{b}^\dagger \hat{b}), \quad (3a)$$

$$\hat{s}_1 = \frac{1}{2}(\hat{a}^\dagger \hat{a} - \hat{b}^\dagger \hat{b}), \quad (3b)$$

$$\hat{s}_2 = \frac{i}{2}(\hat{a}^\dagger \hat{b} - \hat{b}^\dagger \hat{a}), \quad (3c)$$

$$\hat{s}_3 = \frac{1}{2}(\hat{a}^\dagger \hat{b} + \hat{b}^\dagger \hat{a}). \quad (3d)$$

The intensities of light emitted from UP and LP branches can be found from averages $S_\mu(t) = \langle \hat{s}_\mu \rangle$, which satisfy

$$\dot{S}_0 = -\Gamma S_0 + \gamma S_1 - \gamma' S_3, \quad (4a)$$

$$\dot{S}_1 = -(\Gamma + 2\tau_{xc}^{-1})S_1 + \gamma S_0 - \omega_R S_2, \quad (4b)$$

$$\dot{S}_2 = -(\Gamma + \tau_{xc}^{-1})S_2 + \Delta S_3 + \omega_R S_1, \quad (4c)$$

$$\dot{S}_3 = -(\Gamma + \tau_{xc}^{-1})S_3 - \gamma' S_0 - \Delta S_2. \quad (4d)$$

Here $\Gamma = (g_x + g_c + 2\gamma')/2$ and $\gamma = (g_c - g_x)/2$.

To compute the second-order coherence it is convenient to operate with the averages of the normal ordered products $S_{\mu\nu}(t) = \langle : \hat{s}_\mu \hat{s}_\nu : \rangle$ with $\mu, \nu = 0, 1, 2, 3$. The components of the symmetric tensor $S_{\mu\nu}(t)$ obey the equations

$$\begin{aligned} \dot{S}_{11} = & -2\Gamma S_{11} - 2\tau_{xc}^{-1}(2S_{11} - S_{22} - S_{33}) \\ & + 2\gamma S_{01} - 2\omega_R S_{12}, \end{aligned} \quad (5a)$$

$$\dot{S}_{22} = -2\Gamma S_{22} - 2\tau_{xc}^{-1}(S_{22} - S_{11}) + 2\Delta S_{23} + 2\omega_R S_{12}, \quad (5b)$$

$$\dot{S}_{33} = -2\Gamma S_{33} - 2\tau_{xc}^{-1}(S_{33} - S_{11}) - 2\gamma' S_{03} - 2\Delta S_{23}, \quad (5c)$$

$$\begin{aligned} \dot{S}_{01} = & -2\Gamma S_{01} - 2\tau_{xc}^{-1}S_{01} + \gamma(S_{00} + S_{11}) - \gamma' S_{13} - \omega_R S_{02}, \end{aligned} \quad (5d)$$

$$\begin{aligned} \dot{S}_{02} = & -2\Gamma S_{02} - \tau_{xc}^{-1}S_{02} + \gamma S_{12} - \gamma' S_{23} + \Delta S_{03} + \omega_R S_{01}, \end{aligned} \quad (5e)$$

$$\begin{aligned} \dot{S}_{03} = & -2\Gamma S_{03} - \tau_{xc}^{-1}S_{03} + \gamma S_{13} - \gamma'(S_{00} + S_{33}) - \Delta S_{02}, \end{aligned} \quad (5f)$$

$$\begin{aligned} \dot{S}_{12} = & -2\Gamma S_{12} - 5\tau_{xc}^{-1}S_{12} + \gamma S_{02} + \Delta S_{13} + \omega_R(S_{11} - S_{22}), \end{aligned} \quad (5g)$$

$$\begin{aligned} \dot{S}_{13} = & -2\Gamma S_{13} - 5\tau_{xc}^{-1}S_{13} + \gamma S_{03} - \gamma' S_{01} - \Delta S_{12} - \omega_R S_{23}, \end{aligned} \quad (5h)$$

$$\begin{aligned} \dot{S}_{23} = & -2\Gamma S_{23} - 2\tau_{xc}^{-1}S_{23} - \gamma' S_{02} + \Delta(S_{33} - S_{22}) + \omega_R S_{13}. \end{aligned} \quad (5i)$$

Here we omitted the equation for S_{00} , which follows from the identity $S_{00} = S_{11} + S_{22} + S_{33}$, namely, $\dot{S}_{00} = -2\Gamma S_{00} + 2\gamma S_{01} - 2\gamma' S_{03}$. It should be noted that the

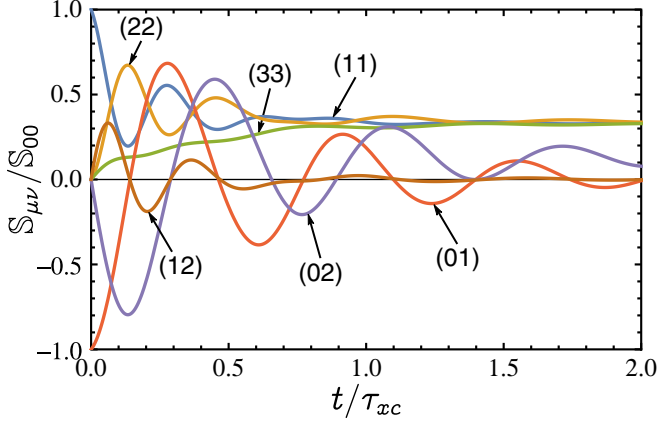


FIG. 2. Showing the time dependencies of normalized nonzero components of the tensor $S_{\mu\nu}$ after excitation of a photon state at $t = 0$. The curves are labeled by indices $(\mu\nu)$. The parameters are $\tau_c/\tau_{xc} = 0.4$, $\tau_x/\tau_{xc} = 1.6$, $\omega_R\tau_{xc} = 10$, and $\Delta = \gamma' = 0$.

Eqs. (4a)–(4d) and (5a)–(5i) are exact consequences of the quantum Liouville equation for the density matrix (1). No quasiclassical assumptions have been made, and one can use these equations for the analysis of evolution of the quantum states of the Rabi oscillator.

In what follows, we consider the pulsed excitation of a photonic state assuming that an ultrashort pulse of light arrives at $t = 0$. For the case of initial coherent photonic state with N photons in average, the initial condition to Eqs. (4a)–(4d) are $S_0(0) = -S_1(0) = N/2$ and $S_2(0) = S_3(0) = 0$. For Eqs. (5a)–(5i) we have $S_{00} = S_{11} = -S_{01} = N^2/4$, and the other components of the tensor are zero. We use the above initial conditions in what follows. We note that in the case of Fock initial state for photonic component, the conditions for the nonzero tensor components should be changed to $S_{00} = S_{11} = -S_{01} = N(N-1)/4$.

In the most experimentally relevant case of photonic excitation the components $S_{0,1,2}(t)$ exhibit decaying Rabi oscillations, and in the absence of stochastic processes ($\tau_{xc}^{-1} = 0$) one has $S_0^2 = S_1^2 + S_2^2 + S_3^2$. This equality is violated in the case of a finite stochastic conversion time τ_{xc} , since the exciton-photon conversion results in decoherence. The evolution of tensor $S_{\mu\nu}$ is more complex and it is characterized by appearance of finite off-diagonal correlations at long times. Even in the simplest case of $\gamma' = 0$ and zero detuning, which is shown in Fig. 2, there are six nonzero components. If exciton and photon decay rates are slow compared to the decoherence rate ($\tau_x, \tau_c \gg \tau_{xc}$), the diagonal S_{11} , S_{22} , and S_{33} components tend to $S_{00}/3$ at long times, indicating the equidistribution of populations of excitons and photons. This tendency is only approximately observed if decay and decoherence rates are of the same order. For the parameters in Fig. 2, we have at long times $S_{11} \rightarrow 0.331S_{00}$, $S_{22} \rightarrow 0.342S_{00}$, and $S_{33} \rightarrow 0.327S_{00}$. Also, at long times, there appear finite off-diagonal correlations, $S_{01} \rightarrow 0.0124S_{00}$, $S_{02} \rightarrow 0.122S_{00}$, and $S_{12} \rightarrow 0.00147S_{00}$.

III. INTENSITY-INTENSITY CORRELATIONS

The Hamiltonian (2) is diagonalized in the basis of LP and UP states with the annihilation operators $\hat{c}_l = \hat{a} \cos \phi -$

$\hat{b} \sin \phi$ and $\hat{c}_u = \hat{a} \sin \phi + \hat{b} \cos \phi$, where the auxiliary angle ϕ is defined by $\tan(2\phi) = \omega_R/\Delta$. The polariton occupation numbers are

$$n_i = \langle \hat{c}_i^\dagger \hat{c}_i \rangle = S_0 \mp S_1 \cos(2\phi) \pm S_3 \sin(2\phi), \quad (6)$$

where the upper and the lower signs correspond to the UP ($i = u$) and the LP ($i = l$) branches, respectively.

There are several second-order correlators that can be experimentally accessed for an exciton-polariton Rabi oscillator. Those of major interest are the UP-LP intensities correlator $g_{ul}(t)$, which can be determined as

$$\begin{aligned} g_{ul} &= \frac{\langle \hat{c}_u^\dagger \hat{c}_l^\dagger \hat{c}_u \hat{c}_l \rangle}{\langle \hat{c}_u^\dagger \hat{c}_u \rangle \langle \hat{c}_l^\dagger \hat{c}_l \rangle} \\ &= \frac{S_{11} \sin^2(2\phi) + S_{22} + S_{33} \cos^2(2\phi) + S_{13} \sin(4\phi)}{S_0^2 - [S_1 \cos(2\phi) - S_3 \sin(2\phi)]^2}, \end{aligned} \quad (7)$$

the second-order coherence for UP and LP,

$$\begin{aligned} g_i^{(2)} &= n_i^{-2} \langle \hat{c}_i^\dagger \hat{c}_i^\dagger \hat{c}_i \hat{c}_i \rangle \\ &= n_i^{-2} [S_{00} + S_{11} \cos^2(2\phi) + S_{33} \sin^2(2\phi) \\ &\quad \mp 2S_{01} \cos(2\phi) \pm 2S_{03} \sin(2\phi) - S_{13} \sin(4\phi)], \end{aligned} \quad (8)$$

with the same convention about the signs as in Eq. (6), and the generalized UP-LP correlator expressed through the previous three correlators,

$$G_{ul} = \frac{g_{ul}^2}{g_u^{(2)} g_l^{(2)}} = \frac{\langle \hat{c}_u^\dagger \hat{c}_l^\dagger \hat{c}_u \hat{c}_l \rangle^2}{\langle \hat{c}_u^\dagger \hat{c}_u \hat{c}_u \hat{c}_u \rangle \langle \hat{c}_l^\dagger \hat{c}_l \hat{c}_l \hat{c}_l \rangle}. \quad (9)$$

The above correlators equate to unity for coherent Rabi oscillations, and their deviation from 1 is a smoking gun of the stochastic exciton-photon conversion processes. Moreover, the effect of decoherence is well pronounced even for rather long conversion times τ_{xc} , as seen in Figs. 3(a)–3(c). The UP-LP correlator (7) shown in Fig. 3(a) decreases below 1, and for short enough τ_{xc} can become close to 2/3. This happens because the exciton-photon conversion process tend to form the equidistribution of particles, and the perfect equidistribution gives $g_{ul} = 2/3$ [10]. However, the equidistribution does not hold with time, because the excitons and photons are removed from the cavity with different rates $\tau_x^{-1} \neq \tau_c^{-1}$. As a result, the time dependence of g_{ul} is nonmonotonous and there is a slow growth of this correlator at long times. The evolution of the polariton second-order coherence (8) is also in general nonmonotonous and exhibits bunching, $g_{u,l}^{(2)} > 1$; see Fig. 3(b). Since both g_{ul} and $g_{u,l}^{(2)}$ increase at long times, it is convenient to consider the combined correlator G_{ul} (9), which tends to a constant value at $t \rightarrow \infty$. This saturation value is 1/4 for short τ_{xc} [10]. All correlators shown in Figs. 3(a)–3(c) reflect the presence of Rabi oscillations in the system, as seen from weak high-frequency modulation of all curves. Since the typical cavity lifetimes are about 10 ps, one can see from Fig. 3 that the color two-photon correlation experiments can reliably detect the stochastic exciton-photon processes with the conversion times as long as nanoseconds.

When τ_{xc} is comparable or larger than τ_c the saturation value of correlator G_{ul} is bigger than 1/4 and it changes with Δ

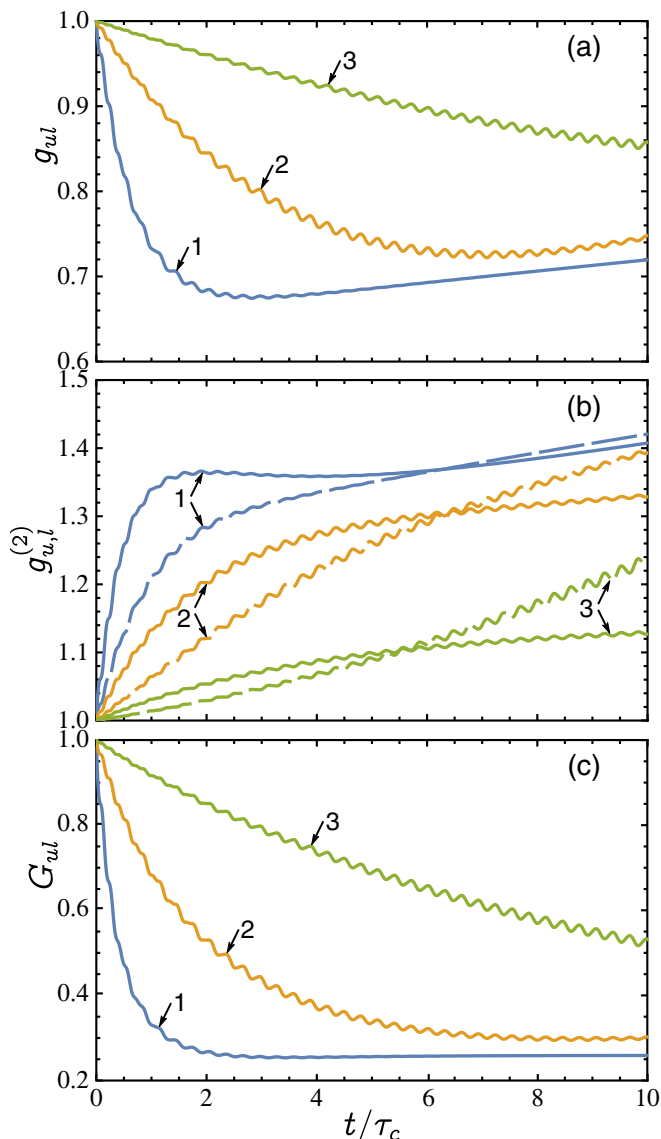


FIG. 3. The time evolution of the correlators (a) g_{ul} , (b) $g_{ul}^{(2)}$ (solid lines) and $g_l^{(2)}$ (dashed lines), and (c) G_{ul} for different exciton-photon conversion times, (1) $\tau_c/\tau_{xc} = 0.5$, (2) $\tau_c/\tau_{xc} = 0.1$, and (3) $\tau_c/\tau_{xc} = 0.02$. We note that in the absence of stochastic processes ($\tau_{xc}^{-1} = 0$) all correlators shown in this figure become time independent and stay equal to 1. The other parameters are $\tau_x = 4\tau_c$, $\omega_R\tau_c = 20$, $\Delta\tau_c = -5$, and $\gamma' = 0$.

and γ' . The dependencies of the asymptotic value G_{ul}^∞ on these parameters are shown in Fig. 4 and they can be used to adjust the experimental conditions for better observation of the effect. Since the parameter γ' governs the difference of dissipation rates of upper and lower polaritons, one can see from Fig. 4 that in the experimentally relevant case of the short upper-polariton

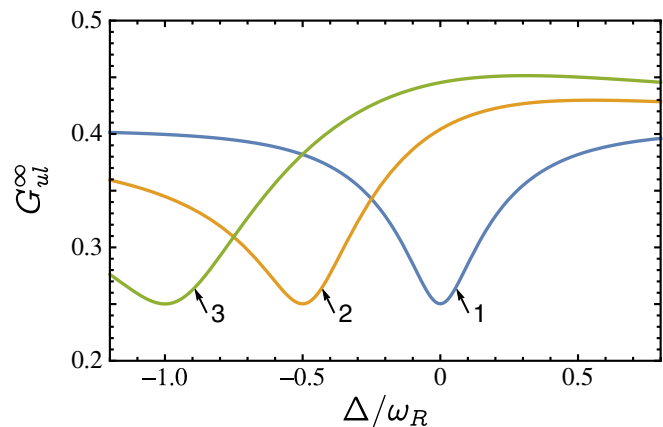


FIG. 4. The asymptotic (long time) value of the correlator G_{ul}^∞ as a function of detuning for $\gamma'/\omega_R = 0$ (curve 1), $\gamma'/\omega_R = 0.01$ (curve 2), and $\gamma'/\omega_R = 0.02$ (curve 3). The other parameters are $\gamma/\omega_R = 0.02$ and $\omega_R\tau_{xc} = 200$.

lifetime it is convenient to use negative detuning between the bare exciton and photon modes, achieving more pronounced decrease of G_{ul} .

IV. CONCLUSION

Two-color intensity-intensity correlation measurements between UP and LP branches in the regime of Rabi oscillations are expected to shed light onto decoherence caused by stochastic exciton-photon conversion processes in the system. The two-color correlator is predicted to go below 1 if the stochastic processes are important. This prediction allows for a relatively simple experimental test of the decoherence of polariton Rabi oscillator and verification of the quantum superposition nature of an exciton-polariton state in the strong coupling regime. We underline that the theory developed here is linear in the pump intensity. The role of exciton-photon correlations in nonlinear optical response of microcavities would constitute a subject of further studies. Finally, we note that a straightforward extension of our formalism permits evaluation of the noise spectra of UP and LP emission intensities. These spectra are also expected to be sensitive to the stochastic exciton-photon conversion.

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