Validity of semiclassical treatment of optical response in cavity systems

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We have investigated the validity of the semiclassical treatment to calculate the third-order nonlinear spectra of cavity systems. Even if applied field is coherent, a quantized effect of a cavity-quasimode manifests itself in the nonlinear spectra in the weak- and strong-coupling regimes, and thus the semiclassical treatment is found to be invalid. One exception exists in the weak-coupling regime without a pure transverse damping.

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I. INTRODUCTION

Optical properties of resonant matter inside a cavity, have been extensively studied because of its potential to achieve large optical nonlinearity, which is necessary to realize optical devices such as the optical switching,[1,2] the optical quantum gates,[3] and so on. At first, optical properties of the cavity systems have been investigated for atoms passing through a cavity, and the field of the cavity quantum electrodynamics (QED) has been explored.[4] An impressive demonstration of an enhanced nonlinear phase shift has been exhibited for Cs atoms in the cavity.[5] More recently, a semiconductor quantum dot (QD) embedded in the cavity has been attracted much attention because of its large oscillator strength and the tractability for fabricating a built-in device.

When applied fields have the same polarization, it has been predicted and measured that the QD can be treated as a two-level system in the optical response.[6–9] If the two-level system is embedded in a lossless cavity, the energy levels have the ladder structure according to the cavity QED, each rung of which consists of two energy levels $E=n\hbar\omega_0 \pm \sqrt{n}g$, where n is the photon number inside the cavity, ω_0 is the resonant frequency of the two-level system and the cavity mode, and g is the coupling constant of the two-level system and the cavity mode. The schematic level structure is shown in Fig. 1.

The energy splitting of the lowest excited rung $(n=1)$ is called the vacuum-Rabi splitting. The vacuum-Rabi splitting can be found in the semiclassical treatment also, and the resulting linear optical spectra agree with that calculated in the cavity-QED treatment.[4] On the other hand, the higher excited rungs, which contribute to the nonlinear processes, can not be derived from the semiclassical theory. Therefore, it is expected that the cavity-QED treatment is necessary to calculate the nonlinear spectra of the cavity systems. For semiconducting nanostructures in the cavity, however, most of theoretical studies on the nonlinear properties have been performed in the semiclassical treatment.[10–12]

In this paper, we clarify the validity of the semiclassical treatment of the third-order nonlinearity. We use the driven Jaynes-Cumings (JC) model to describe the interaction of the QD and the cavity quasimode fed from the outside coherent field. Both longitudinal- and pure transverse-damping effects are included by employing the master equation. In general, the semiclassical treatment is invalid to calculate the nonlinear spectra except for the weak-coupling regime without the pure transverse damping.

This paper is organized as follows: In Sec. II, the driven JC Hamiltonian and the master equation are presented. The procedure to calculate the third-order nonlinear field is described in the cavity-QED treatment and its semiclassical approximation. In Sec. III, we compare the spectra obtained from the cavity-QED and semiclassical treatments in the weak- and strong-coupling regimes. A summary is given in Sec. IV.

II. THE TWO-LEVEL SYSTEM IN A CAVITY

Optical response of the two-level system in the cavity can be well described by the driven JC model.[4] The driven JC model consists of cavity-quasimode creation and annihilation operators $a^\dagger$ and a connecting applied coherent field $\mathcal{E}e^{-i\omega t}$ with coupling constant $\sqrt{\Gamma}$. The phenomenological description has been derived from a more fundamental approach,[13,14] in which electromagnetic fields extending inside and outside cavity are quantized.

Let us consider a two-side cavity, inside which the two-level system interacts with the cavity-quasimode photons

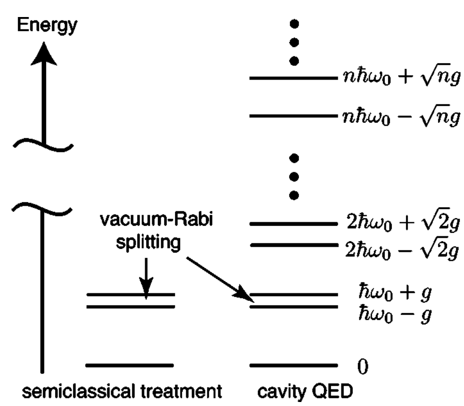

FIG. 1. Energy level structures of the coupled modes of a two-level system and a cavity mode. The vacuum-Rabi splitting appears both in the semiclassical and cavity-QED treatments.

with the coupling constant g. The cavity-quasimode energy of interest is denoted by $\hbar\omega_c$, and the excitation energy of the two-level system is denoted by $\hbar\omega_a$. Then the driven JC Hamiltonian is given by

$$H = \tfrac{1}{2}\hbar\omega_a\sigma_z + \hbar\omega_c a^\dagger a + i\hbar g(a^\dagger\sigma_- - \sigma_+ a) + i\hbar\sqrt{\Gamma}\mathcal{E}(ae^{i\omega t} - a^\dagger e^{-i\omega t}), \tag{1}$$

where $\sigma_\pm$, σ_z are pseudospin operators for the two-level system.

Decay processes of the excited states are usually characterized by the longitudinal (γ_1) and pure transverse (γ_2) damping constants. Photons inside the cavity decay to the outside because of the imperfection of the cavity mirrors, and the corresponding decay constant is given by Γ. The Q factor of the cavity is related to Γ as $Q=\omega_c/2\Gamma$. In the presence of these decay processes, a density matrix ρ for the two-level system and the cavity quasimode satisfies the master equation

$$\frac{d\rho}{dt} = \frac{1}{i\hbar}[H,\rho] + \frac{\gamma_1}{2}(2\sigma_-\rho\sigma_+ - \sigma_+\sigma_-\rho - \rho\sigma_+\sigma_-) + \frac{\gamma_2}{2}(\sigma_z\rho\sigma_z - \rho) + \Gamma(2a\rho a^\dagger - a^\dagger a\rho - \rho a^\dagger a). \tag{2}$$

The driven JC Hamiltonian (1) and the master Eq. (2) determine optical response of the two-level system embedded in the two-side cavity. The response field E is calculated from the input-output theory[15] as

$$E = \sqrt{\Gamma}\langle a\rangle, \tag{3}$$

with $\langle a\rangle = \mathrm{Tr}(\rho a)$.

A. The cavity-QED treatment

Here, we present a quantum treatment of the cavity quasimode in the perturbation with respect to $\mathcal{E}$. In the following calculations, we consider the degenerate case: $\omega_c=\omega_a\equiv\omega_0$. In the steady-state condition, the equations of motion of $\langle a\rangle$, $\langle\sigma_-\rangle$, $\langle\sigma_z\rangle$, and $\langle a\sigma_z\rangle$ are given by

$$iA\langle a\rangle + g\langle\sigma_-\rangle - \sqrt{\Gamma}\mathcal{E} = 0, \tag{4}$$

$$iB\langle\sigma_-\rangle + g\langle a\sigma_z\rangle = 0, \tag{5}$$

$$2g\langle a^\dagger\sigma_-\rangle + 2g\langle\sigma_+ a\rangle + \gamma_1\langle\sigma_z\rangle + \gamma_1 = 0, \tag{6}$$

$$iC\langle a\sigma_z\rangle - 2g\langle a^\dagger a\sigma_-\rangle - 2g\langle aa\sigma_+\rangle - g\langle\sigma_-\rangle - \sqrt{\Gamma}\mathcal{E}\langle\sigma_z\rangle - \gamma_1\langle a\rangle/2 = 0, \tag{7}$$

respectively, with $A=\Delta\omega+i\Gamma$, $B=\Delta\omega+i(\gamma_1/2+\gamma_2)$, and $C=\Delta\omega+i\Gamma+i\gamma_1$ where $\Delta\omega=\omega-\omega_0$.

The zeroth order solution $\langle\sigma_z\rangle^{(0)}=-1$ is obtained from Eq. (6). The first-order solutions are calculated from Eqs. (4), (5), and (7) as follows:

$$\langle\sigma_-\rangle^{(1)} = \frac{-g\sqrt{\Gamma}}{AB-g^2}\mathcal{E}, \tag{8}$$

$$\langle a\rangle^{(1)} = -i\frac{\sqrt{\Gamma}B}{AB-g^2}\mathcal{E}. \tag{9}$$

It is noted that the solutions $AB-g^2=0$ provide the complex vacuum-Rabi frequencies

$$\lambda_\pm = \omega_0 - \frac{i}{4}(2\Gamma + \gamma_1 + 2\gamma_2) \pm \sqrt{g^2 - (2\Gamma - \gamma_1 - 2\gamma_2)^2/16}. \tag{10}$$

The real and imaginary parts of $\lambda_\pm$ correspond to the resonant frequency and the spectral width of the linear transmission spectra, respectively. For $4g\leqslant 2\Gamma-\gamma_1-2\gamma_2$, we have $\mathrm{Re}[\lambda_+]=\mathrm{Re}[\lambda_-]=\omega_0$ (weak-coupling regime). For $4g>2\Gamma-\gamma_1-2\gamma_2$, on the other hand, the real parts have different values, which lead to the vacuum-Rabi splitting (strong-coupling regime).

The equations of motion to obtain the second-order solutions are written as

$$\begin{pmatrix} \gamma_1 & 2g & 2g & 0 & 0 & 0 \\ g/2 & -\xi & 0 & 0 & 0 & -g \\ g/2 & 0 & -\xi & 0 & 0 & -g \\ 0 & 0 & 0 & iD & -g & 0 \\ 0 & 0 & 0 & 2g & iE & 0 \\ 0 & g & g & 0 & 0 & -2\Gamma \end{pmatrix} \begin{pmatrix} \langle\sigma_z\rangle^{(2)} \\ \langle a^\dagger\sigma_-\rangle^{(2)} \\ \langle a\sigma_+\rangle^{(2)} \\ \langle a\sigma_-\rangle^{(2)} \\ \langle aa\rangle^{(2)} \\ \langle a^\dagger a\rangle^{(2)} \end{pmatrix} = \sqrt{\Gamma}\mathcal{E} \begin{pmatrix} 0 \\ \langle\sigma_-\rangle^{(1)} \\ \langle\sigma_+\rangle^{(1)} \\ \langle\sigma_-\rangle^{(1)} \\ 2\langle a\rangle^{(1)} \\ \langle a\rangle^{(1)} + \langle a^\dagger\rangle^{(1)} \end{pmatrix}, \tag{11}$$

with $D=2\Delta\omega+i\Gamma+i(\gamma_1/2+\gamma_2)$, $E=2\Delta\omega+2i\Gamma$, and $\xi=\Gamma+(\gamma_1/2+\gamma_2)$.

A simultaneous equation providing the third-order solutions is given by

$$\begin{pmatrix} iA & g & 0 & 0 & 0 & 0 \\ 0 & iB & g & 0 & 0 & 0 \\ -\gamma_1 & -g & iC & -2g & -2g & 0 \\ g/2 & 0 & g/2 & iF & 0 & -g \\ g & 0 & g & 0 & iF & -g \\ 0 & 0 & 0 & 2g & g & iG \end{pmatrix} \begin{pmatrix} \langle a\rangle^{(3)} \\ \langle\sigma_-\rangle^{(3)} \\ \langle a\sigma_z\rangle^{(3)} \\ \langle a^\dagger a\sigma_-\rangle^{(3)} \\ \langle aa\sigma_+\rangle^{(3)} \\ \langle a^\dagger aa\rangle^{(3)} \end{pmatrix} = \sqrt{\Gamma}\mathcal{E} \begin{pmatrix} 0 \\ 0 \\ \langle\sigma_z\rangle^{(2)} \\ \langle a\sigma_-\rangle^{(2)} + \langle a^\dagger\sigma_-\rangle^{(2)} \\ 2\langle a\sigma_+\rangle^{(2)} \\ \langle aa\rangle^{(2)} + 2\langle a^\dagger a\rangle^{(2)} \end{pmatrix}. \tag{12}$$

with $F=\Delta\omega+2i\Gamma+i(\gamma_1/2+\gamma_2)$ and $G=\Delta\omega+3i\Gamma$. The third-order nonlinear response $E^{(3)}=\sqrt{\Gamma}\langle a\rangle^{(3)}$ is calculated from the simultaneous Eqs. (11) and (12) numerically.

B. The semiclassical treatment

The semiclassical treatment corresponds to the approximation of the cavity-QED treatment, where the operator a is replaced by its mean value, for instance, $\langle a\sigma_z\rangle \approx \langle a\rangle\langle\sigma_z\rangle$. Applying the approximation to Eqs. (4)–(6), we obtain the equations of motion in a closed form

$$\langle a\rangle = i\frac{g}{A}\langle\sigma_-\rangle - i\frac{\sqrt{\Gamma}}{A}\mathcal{E}, \tag{13}$$

$$\langle\sigma_-\rangle = i\frac{g}{B}\langle a\rangle\langle\sigma_z\rangle, \tag{14}$$

$$\gamma_1\langle\sigma_z\rangle = -\gamma_1 - 2g[\langle a^\dagger\rangle\langle\sigma_-\rangle + \langle a\rangle\langle\sigma_+\rangle]. \tag{15}$$

The zeroth order of $\langle\sigma_z\rangle$ is given by $\langle\sigma_z\rangle^{(0)}=-1$ from Eq. (15). The calculated $\langle a\rangle^{(1)}$ and $\langle\sigma_-\rangle^{(1)}$ have the same form as Eqs. (8) and (9), respectively. Namely, the semiclassical approximation leads to the same result as that in the cavity-QED treatment in the linear-response regime.[4]

To derive the third-order nonlinear spectra we have

$$\langle\sigma_z\rangle^{(2)} = \frac{\gamma_1 + 2\gamma_2}{\gamma_1}\frac{2g^2\Gamma}{|AB - g^2|^2}\mathcal{E}^2, \tag{16}$$

by substituting Eqs. (8) and (9) into Eq. (15). After getting $\langle\sigma_-\rangle^{(3)}$, we have the third-order response field $E^{(3)}$

$$E^{(3)} = \sqrt{\Gamma}\langle a\rangle^{(3)} = i\frac{\gamma_1 + 2\gamma_2}{\gamma_1}\frac{2g^4\Gamma^2}{A(AB - g^2)|AB - g^2|^2}\mathcal{E}^3. \tag{17}$$

It has been demonstrated that the present semiclassical approximation of the cavity-QED treatment agrees with the conventional semiclassical approach on the basis of the Maxwell equations and boundary conditions at cavity mirrors.[16] The analytic demonstration has been performed for a high-Q cavity, in which the cavity quasimode is well defined, and the phenomenological parameter Γ has been related to the transmission coefficient t of the mirrors as $\sqrt{\Gamma}=-i\sqrt{c/2L}t$ where L is the length of the cavity.

Figure 2 shows the third-order nonlinearities calculated from various approaches for a low-Q cavity. The parameters in the following calculations are selected for the interface GaAs QDs as the two-level system. The QDs have large transition-dipole moment ranging from 50 to 100 D.[17,18] The typical γ_2 of the interface QD at $T=12$ K is $\gamma_2 \approx 16-27$ μeV.[9] The parameters are selected as $\omega_0=1.65$ eV,[19] $\gamma_1=8$ μeV,[19] and $\gamma_2=16$ μeV. The coupling constant is given by $\hbar g = P\sqrt{4\pi\hbar\omega_0/SL}$, where P is a transition-dipole moment and S is a cross section of the applied laser field. The energy of the lowest cavity quasimode is adjusted to the excitation energy of the QD, i.e., $\omega_0=L/c\pi$. Then the coupling constant is rewritten as $\hbar g = 2(P/a)(\hbar\omega_0/\sqrt{\hbar c})$. When the Gaussian waist w_0 of the applied laser field is 10 μm, the coupling energy becomes 0.07–0.14 meV for $P=50-100$ D, where I have used the relation $S=\pi w_0^2/4$.[4] Here, the coupling constant is fixed at $g=0.1$ meV.

The intensity is normalized by the maximum intensity $|E_0^{(3)}|$ for the QD without the cavity. Solid curves indicated by

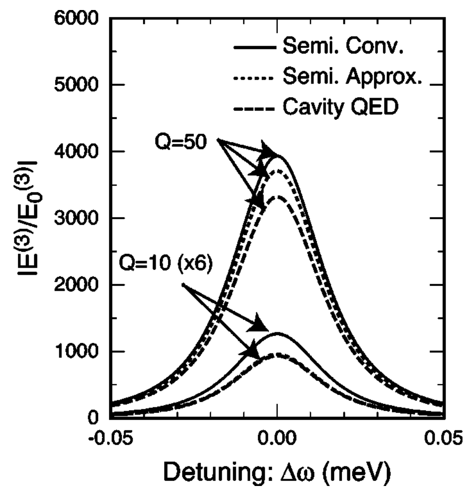

FIG. 2. The third-order nonlinear spectra $|E^{(3)}|$ for the two-level system in the cavity with $Q=50$ and 10 as a function of the detuning $\Delta\omega$. The intensity is normalized by the maximum one $|E_0^{(3)}|$ without the cavity. They are calculated in the semiclassical approach on the basis of the Maxwell equations (solid curves), the semiclassical approximation of the cavity QED (dotted curves), and the cavity QED treatment (dashed curves).

"semi. conv." show the results of the conventional semiclassical approach on the basis of the Maxwell equations, and dotted curves indicated by "semi. approx." show the results of Eq. (17), namely, the semiclassical approximation of the cavity QED. It is found from these results that the driven JC Hamiltonian describes cavity systems relatively well even in the low-Q regime. The dashed curves indicated by "cavity QED" shows the results of the cavity-QED treatment.

III. COMPARISON OF THE CAVITY-QED AND THE SEMICLASSICAL TREATMENTS

A. The weak-coupling regime

In the case of the weak-coupling regime ($\Gamma \gg g$), the cavity quasimode operator a can be eliminated adiabatically,[20] in which a is approximated as

$$a = i\frac{g}{A}\sigma_- - i\frac{\sqrt{\Gamma}}{A}\mathcal{E}. \tag{18}$$

In the adiabatic-elimination procedure, operator a is substituted into the equations of motion for $\langle\sigma_-\rangle$ and $\langle\sigma_z\rangle$, and we obtain the equations of motion of the pseudospin operators in a closed form.

Substituting Eq. (18) into Eq. (5), we have

$$\langle\sigma_-\rangle = \frac{g\sqrt{\Gamma}\mathcal{E}}{AB - g^2}\langle\sigma_z\rangle. \tag{19}$$

The $\langle\sigma_z\rangle^{(2)}$ is calculated from Eqs. (18) and (6)

$$\langle\sigma_z\rangle^{(2)} = 2g^2\Gamma\frac{\delta + 2\gamma_2|A|^2}{\delta|AB - g^2|^2}\mathcal{E}^2, \tag{20}$$

with $\delta=2g^2\Gamma + \gamma_1|A|^2$. Then $\langle\sigma_-\rangle^{(3)}$ is obtained from Eq. (19), and $\langle a\rangle^{(3)}$ is calculated. Thus the $E^{(3)}$ is given by

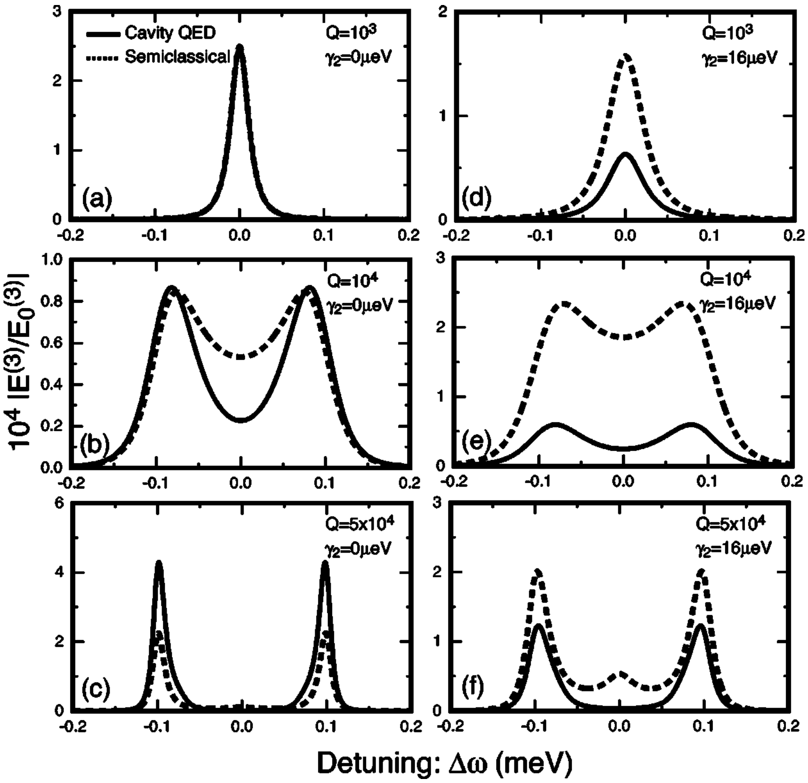

FIG. 3. Calculated spectra of $|E^{(3)}|$ as a function of the detuning $\Delta\omega$ for various Q factors.

$$E^{(3)} = i\frac{\delta + 2\gamma_2|A|^2}{\delta}\frac{2g^4\Gamma^2}{A(AB-g^2)|AB-g^2|^2}\mathcal{E}^3. \qquad (21)$$

The expressions of $E^{(3)}$ in the semiclassical approximation (17) and the cavity-QED treatment (21), are different. For extremely low-Q cavity, however, the cavity-QED treatment should lead to the same result obtained in the semiclassical approach. In fact, Eq. (21) is approximated as the semiclassical result (17) in the extremely weak-coupling regime satisfying $\gamma_1 \gg g^2/\Gamma$.

In Fig. 2 of the previous section, we compare the third-order nonlinear spectra calculated from the cavity QED (dashed curves) and semiclassical treatments (dotted curves) for extremely low-Q cavity ($Q=10$ and 50). The difference between the two approaches becomes much smaller with decrease of the Q factor. Thus the cavity-QED effect disappears in the extremely weak-coupling regime.

For $\gamma_2=0$ in the weak-coupling regime, the nonlinear spectra obtained from the cavity-QED and semiclassical treatments become in agreement with each other. In other words, the semiclassical treatment gives the correct nonlinear field in the weak-coupling regime provided that the pure transverse damping is negligible.

Let us compare the results of the cavity-QED and semiclassical treatments numerically. The results of the cavity-QED are obtained by solving Eqs. (11) and (12). The coupling energy is fixed at $g=0.1$ meV, and the weak- and strong-coupling regimes are attained by changing the Q factor. Figure 3 shows the intensity of the third-order nonlinear spectra in the weak- and strong-coupling regimes. The calculations are performed in the cavity-QED treatment (solid curves) and the semiclassical approximation (dotted curves) given in Eq. (17). The left column shows the results for $\gamma_2=0$, in which the semiclassical treatment has been demonstrated to be justified in the weak-coupling regime. In fact, the semiclassical approximation provides the same result as that in the cavity-QED treatment for $Q=10^3$ and $\gamma_2=0$ [see Fig. 3(a)]. The right column shows the nonlinear spectra for $\gamma_2=16$ μeV. As is demonstrated, the semiclassical approximation is found to be invalid even in the weak-coupling regime [see Fig. 3(d)]. The semiclassical approximation overestimates the third-order nonlinearity considerably for $\gamma_2 \neq 0$.

B. Strong-coupling regime

In the strong-coupling regime, intensity of the nonlinear spectra in the semiclassical approximation is different from that in the cavity-QED treatment both for $\gamma_2=0$ and 16 μeV [see Figs. 3(b), 3(c), 3(e), and 3(f)]. In addition to the quantitative incorrectness, semiclassical treatment has some qualitative errors in the strong-coupling regime. One of the errors is the enhancement of optical nonlinearity due to the pure dephasing for $Q=10^4$ [see Figs. 1(b) and 1(e)]; the intensity of $E^{(3)}$ for $\gamma_2=16$ μeV becomes larger than that for $\gamma_2=0$. This tendency is not found in the cavity-QED treatment at all. In the semiclassical result for $\gamma_2 \neq 0$, furthermore, there exists a peak structure at $\Delta\omega=0$, and appreciable nonlinear intensity appears between the Rabi-splitting peaks in the extremely strong-coupling regime [see Fig. 1(f)]. In the cavity-QED treatment, the nonlinear intensity is quite small between the Rabi-splitting peaks.

IV. SUMMARY AND CONCLUSION

In summary, the validity of the semiclassical approximation of the optical response, has been investigated for the two-level system embedded in the cavity, by comparing the results of the cavity-QED treatment. Although there is no difference between semiclassical and cavity-QED treatments in the linear response, cavity-QED effect appears in the nonlinear response.

Except for the extremely weak-coupling regime with $\gamma_1 \gg g^2/\Gamma$, the semiclassical approximation is invalid in the nonlinear response of the cavity systems in the weak- (with $\gamma_2 \neq 0$) and strong-coupling regimes. This fact can be expected easily in the strong-coupling regime because of the well-defined excited rungs of the energy ladder structure in the cavity QED. Except for the lowest excited rung (or vacuum-Rabi splitting), the energy ladder structure is not found in the semiclassical approximation.

In the strong-coupling regime, the semiclassical approximation leads to incorrect results qualitatively and quantitatively. In some cases of the semiclassical treatment, the pure transverse damping enhances the intensity of the third-order nonlinear spectra considerably [see Figs. 3(b) and 3(e)]. In addition, appreciable intensity of the nonlinear spectra is found between the Rabi-splitting peaks, which has a peak structure at $\Delta\omega = 0$ [see Fig. 3(f)]. These results are not found in the cavity-QED treatment.

The vacuum-Rabi splitting disappears in the weak-coupling regime. However, the discrepancy of the nonlinear spectra between the semiclassical and cavity-QED treatments is appreciable in the weak-coupling regime with $\gamma_2 \neq 0$. The discrepancy has been analytically demonstrated and numerically shown in Fig. 3(d). In the absence of the pure transverse damping, however, the semiclassical approximation has been demonstrated to be valid [see also Fig. 3(a)].

The validity of the optical nonlinearity of the cavity systems, has been studied in the two-level system. However, the present results would be also applicable for other low-dimensional systems such as quantum well.

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