NON-CLASSICAL LIGHT SCATTERED BY LASER-PUMPED MOLECULES POSSESSING PERMANENT DIPOLES

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Abstract

Squeezing in resonance fluorescence processes of a laser-pumped molecule possessing permanent dipoles is shown. A weak low-frequency coherent field pumps the sample near resonance with the dynamically Stark-splitting states induced by a second stronger laser field driving the main two-level transition. The fluorescence spectrum consists of three coherently scattered spectral lines and up to nine incoherently scattered spectral lines. Compared with a similar problem yet in the absence of permanent dipoles, aditional squeezing regions are found.

1. Introduction

The resonance fluorescence phenomenon has attracted great attention in the last decades; the Mollow spectrum is a clear example from this point of view. The potential applications with artificial atomic systems, such as semiconductor quantum dots, have renewed interest in this topic. Furthermore, another important nonclassical feature of the resonance fluorescence spectrum is the squeezing of the field quadratures of a two-level system, which was theoretically adressed by Walls and Zoller [1] and later experimentally verified. Owing to potential appplications in high-precisioon measurements, such as graviational wave detection or quantum computing, the squeezing of the fluorescent field has been widely studied in two- and three-level atoms driven by a laser. In connection with the developement of quantum informatics, squeezed states of the radiation field have been recongized as crucial resources for continous variable quantum information processing. Therefore, the issue of generation of fields with enhanced squeezing is still an important topic [1, 2].

Semiconductor quantum dots are considered attractive candidates for many optoelectronic applications as single-photon sources, lasers, and computational building blocks of a quantum computer. From the fundamental point of view, an outstanding application of the artificial atom model where excitation can be considered as a two-level system is the developement of quantum optics experiments in solid state physics. The emission of photons with a sub-poissonian photon statistics, as well as antibunched photons, has been already shown [3]; these achievements could be used for quantum cryptography [4, 5]. Rabi oscillations are another attractive phenomenon, which can be observed when a single quantum dot is strongly driven by a laser under quasi-resonant excitation [6, 7]. The coupling between the excitonic two-level system and an optical cavity mode leads to the cavity quantum electrodynamics research field in solid state systems, similarly to single atoms in cavity [8]. As a consequence, another approach to coherently control the exciton quantum bit in a single quantum dot and generate non-classical states of light is to benefit from the strong coupling of a quantum dot two-level system with an optical microcavity and from the resulting quantum anharmonicty of the energy structure of the quantum dot–cavity

system [9].
Here, we study a two-level quantum emitter which is pumped by two coherent laser fields and scatters photons coherently or incoherently via the surrounding vacuum modes of the electromagnetic field reservoir. The first low-frequency coherent field pumps the sample near resonance with the dynamically Stark-splitting states induced by a second stronger laser field driving the main two-level transition. Under these conditions, we have found that the fluorescence spectrum consists of three coherently scattered spectral lines and up to nine incoherently scattered spectral lines. In particular, we have shown aditional squeezing regions found in resonance fluorescence processes in comparison with similar problem yet in the absence of permanent dipoles.

This paper is organized as follows. In Section 2, we describe the analytic model and analyze the relevant equations of motion using the system's master equation. In Section 3, we compute the resonance fluorescence spectrum, while in Section 4 we give the squeezed spectrum, respectively. We finalize the paper with a summary given in Section 5.

2. Theoretical Framework

Let us consider a two-level system possessing permanent dipoles and interacting with two external coherent laser fields. The first laser is near resonance with the transition frequency of the two-level sample, while the second laser is close to resonance with the dressed-frequency splitting due to the first laser. In the dipole approximation, the Hamiltonian describing this model, in a frame rotating at the first laser frequency ω_L, is as follows [10, 11]:

$$H = \sum_k \hbar\omega_k a_k^\dagger a_k + \hbar\omega_0 S_z + \hbar\Omega_1\left(S^+ e^{-i\omega_L t} + S^- e^{i\omega_L t}\right) + \hbar\Omega_2(S^+ + S^-)\cos(\omega t) +$$
$$+\hbar G S_z \cos(\omega t) + \hbar G_1 S_z \cos(\omega_L t) + i\,\hbar \sum_k \left(\vec{g}_k \cdot \vec{d}\right)\left\{a_k^\dagger S^- - a_k S^+\right\}, \qquad (1)$$

In Hamiltonian (1), the first four components are the free energies of the environmental electromagnetic vacuum modes and molecular subsystems together with the laser-molecule interaction Hamiltonian, respectively. Here, $\Omega_1 \equiv \Omega = dE_1/(2\,\hbar)$ is the corresponding Rabi frequency with $d \equiv d_{21} = d_{12}$ being the transition dipole moment while E_1 is the amplitude of the first laser field. The fifth term accounts for the second laser interacting at frequency ω and amplitude E_2 with the molecular system due to the presence of permanent dipoles incorporated in G, i.e., $G = (d_{22} - d_{11})E_2/\hbar$, while the sixth terms is due to the interaction of the first laser with the permanent dipoles. The last term describes the interaction of molecular subsystems with the environmental vacuum modes of the electromagnetic field reservoir. Further, $\vec{g} = \sqrt{2\,\pi\hbar\omega_k/V}\,\vec{e}_\lambda$ is the molecule–vacuum coupling strength with $\vec{e}_\lambda$ being the photon polarization vector and $\lambda \in \{1,2\}$, whereas V is the quantization volume; $\Delta = \omega_{21} - \omega_L$ is the laser field detuning from the molecular transition frequency ω_{21}. The molecule bare-state operators $S^+ = |2\rangle\langle 1|$ and $S^- = [S^+]^\dagger$ obey the commutation relations $[S^+, S^-] = 2S_z$ and $[S_z, S^\pm] = \pm S^\pm$. Here, $S_z = (|2\rangle\langle 2| - |1\rangle\langle 1|)/2$ is the bare-state inversion operator; $|2\rangle$ and $|1\rangle$ are the excited and ground state of the molecule, respectively, while $a_k^\dagger$ and a_k are the creation and annihilation operators of the k_{th} electromagnetic field mode and satisfy standard bosonic commutation relations, namely, $\left[a_k, a_k^\dagger\right] = \delta_{kk'}$ and $[a_k, a_{k'}] = \left[a_k^\dagger, a_{k'}^\dagger\right] = 0$. In the following, we will neglect the fast oscillating terms and consider a regime where the generalized Rabi frequency $\bar{\Omega} = \sqrt{\Omega^2 + (\Delta/2)^2}$ is larger than the single-molecule spontaneous decay rate, as well as the coupling due to permanent dipoles, i.e., $\bar{\Omega} \gg \gamma$ and $\bar{\Omega} \gg G$. In this case, it is more convenient to describe our system in the semiclassical laser–molecule dressed state picture due to the first applied laser [2]:

$$|2\rangle = -\sin\theta\,|\bar{1}\rangle + \cos\theta\,|\bar{2}\rangle, \text{ and } |1\rangle = \cos\theta\,|\bar{1}\rangle + \sin\theta|\bar{2}\rangle, \tag{2}$$

with $\tan 2\theta = \frac{2\bar{\Omega}}{\Delta}$. Here, $|\bar{2}\rangle$ and $|\bar{1}\rangle$ are the corresponding upper and lower dressed states, respectively. Applying the dressed-state transformations in Hamiltonian (1), one arrives at the following Hamiltonian represented in a rotating frame also at the second laser field frequency, i.e., ω:

$$H = \sum_k \hbar(\omega_k - \omega_L)a_k^\dagger a_k + \hbar\bar{\Delta}R_z - \hbar\bar{G}(R^+ + R^-) +$$
$$+ i\sum_k (\vec{g}_k \cdot \vec{d})\left\{a_k^\dagger\left(\frac{1}{2}\sin 2\theta\, R_z + \cos^2\theta R^- e^{-i\,\omega t} - \sin^2\theta R^+ e^{i\,\omega t}\right) - H.C\right\}, \tag{3}$$

where $\bar{\Delta} \equiv \bar{\Omega} - \omega/2$, $\bar{G} = \frac{1}{4}G\sin 2\,\theta$, $R^+ = |\bar{2}\rangle\langle\bar{1}|$, $R^- = |\bar{1}\rangle\langle\bar{2}|$, $\bar{R}_z = |\bar{2}\rangle\langle\bar{2}| - |\bar{1}\rangle\langle\bar{1}|$. Here, we have assumed that $\omega \gg \bar{G}$ and performed the rotating wave approximation with respect to ω [10, 11]. Introducing Hamiltonian (3) in the Heisenberg equation describing the mean quantum evolution of any molecular operator Q, we obtain

$$\frac{d}{dt}\langle Q(t)\rangle = \frac{i}{\hbar}\langle[\hbar\bar{\Delta}R_z - \hbar\bar{G}(R^+ + R^-), Q]\rangle -$$
$$-\sum_k \frac{(\vec{g}_k \cdot \vec{d})}{\hbar}\left\{\langle a_k^\dagger\left[\frac{1}{2}\sin 2\,\theta\, R_z + \cos^2\theta R^- e^{-i\,\omega t} - \sin^2\theta R^+ e^{+i\,\omega t}\right], Q\rangle - H.C\right\}. \tag{4}$$

One can apply the double dressed-state formalism in order to obtain further information on our system, namely,

$$|\bar{2}\rangle = \sin\bar{\theta}\,|\tilde{1}\rangle + \cos\bar{\theta}|\tilde{2}\rangle\,,\ |\bar{1}\rangle = \cos\bar{\theta}\,|\tilde{1}\rangle - \sin\bar{\theta}|\tilde{2}\rangle. \tag{5}$$

Introducing (5) in Hamiltonian (4) and eliminating the degrees of freedom related with the environmental vacuum modes in the Born–Markov approximations, one arrives at the following double dressed master equation:

$$\frac{d}{dt}\langle Q(t)\rangle = i\bar{G}_R\langle[\tilde{R}_z, Q]\rangle - \bar{\Gamma}_0\{\langle\tilde{R}_z[\tilde{R}_z, Q]\rangle + \langle[Q, \tilde{R}_z]\tilde{R}_z\rangle\} -$$
$$-\bar{\Gamma}_+\{\langle\tilde{R}^+[\tilde{R}^-, Q]\rangle + \langle[Q, \tilde{R}^+]\tilde{R}^-\rangle\} - \bar{\Gamma}_-\{\langle\tilde{R}^-[\tilde{R}^+, Q]\rangle + \langle[Q, \tilde{R}^-]\tilde{R}^+\rangle\}. \tag{6}$$

here,

$$\bar{\Gamma}_0 = \frac{1}{4}\gamma(\omega_L)\sin^2 2\,\theta\cos^2 2\bar{\theta} + \frac{1}{4}\gamma(\omega_L + \omega)\sin^2 2\bar{\theta}\cos^4\theta + \frac{1}{4}\gamma(\omega_L - \omega)\sin^2 2\bar{\theta}\sin^4\theta,$$

$$\bar{\Gamma}_+ = \frac{1}{4}\gamma(\omega_L + 2\bar{G}_R)\sin^2 2\bar{\theta}\sin^2 2\,\theta +$$
$$+\frac{1}{4}\gamma(\omega_L + \omega + 2\bar{G}_R)\cos^4\bar{\theta}\cos^4\theta + \frac{1}{4}\gamma(\omega_L - \omega + 2\bar{G}_R)\sin^4\theta\sin^4\bar{\theta},$$

$$\bar{\Gamma}_- = \frac{1}{4}\gamma(\omega_L - 2\bar{G}_R)\sin^2 2\,\theta\sin^2 2\bar{\theta} +$$

$$+\frac{1}{4}\gamma(\omega_L + \omega - 2\bar{G}_R)\cos^4\theta\sin^4\bar{\theta} + \frac{1}{4}\gamma(\omega_L - \omega - 2\bar{G}_R)\sin^4\theta\cos^4\bar{\theta},$$

with $\gamma(\mathrm{x}) = \frac{2\mathrm{d}^2 x^3}{3\,\hbar c^3}$, $\bar{G}_R = \sqrt{\bar{\Delta}^2 + \bar{G}^2}$, $\bar{\Delta} = \bar{\Omega} - \omega/2$, $\cot 2\bar{\theta} = \frac{\bar{\Delta}}{\bar{G}}$. The new operators, i.e., $\tilde{R}^+ = |\tilde{2}\rangle\langle\tilde{1}|$, $\tilde{R}^- = [\tilde{R}^+]^\dagger$ and $\tilde{R}_z = |\tilde{2}\rangle\langle\tilde{2}| - |\tilde{1}\rangle\langle\tilde{1}|$, are operating in the double dressed state picture obeying the following commutation relations: $[\tilde{R}^+, \tilde{R}^-] = 2\tilde{R}_z$ and $[\tilde{R}_z, \tilde{R}^\pm] = \pm 2\tilde{R}^\pm$. The master equation (6) contains slowly varying terms in the spontaneous emission damping; that is, we have assumed that $\bar{G}_R \gg \gamma(\omega_{21})$, with $\gamma(\omega_{21}) = \frac{2d^2\omega_{21}^3}{3\,\hbar c^3}$ being the single-molecule spontaneous decay rate corresponding to the double dressed-state frequency ω_{21}.
With the help of the master equation (6), we obtain the Bloch system of differential equations, describing our sample, which is solved first in the stationary case, i.e., we consider that $\frac{d}{dt}\langle\tilde{R}^-(t)\rangle = 0$; $\frac{d}{dt}\langle\tilde{R}^+(t)\rangle = 0$; $\frac{d}{dt}\langle\tilde{R}_z(t)\rangle = 0$ in

$$\begin{cases} \frac{d}{dt}\langle\tilde{R}_z(t)\rangle = -2(\bar{\Gamma}_+ + \bar{\Gamma}_-)\langle\tilde{R}_z\rangle + 2(\bar{\Gamma}_- - \bar{\Gamma}_+), \\ \frac{d}{dt}\langle\tilde{R}^+(t)\rangle = (2i\bar{G}_R - 4\bar{\Gamma}_0 - \bar{\Gamma}_+ - \bar{\Gamma}_-)\langle\tilde{R}^+\rangle, \\ \frac{d}{dt}\langle\tilde{R}^-(t)\rangle = -(2i\bar{G}_R + 4\bar{\Gamma}_0 + \bar{\Gamma}_+ + \bar{\Gamma}_-)\langle\tilde{R}^-\rangle, \end{cases} \tag{7}$$

to arrive at: $\langle\tilde{R}_z\rangle_s = \frac{\bar{\Gamma}_- - \bar{\Gamma}_+}{\bar{\Gamma}_- + \bar{\Gamma}_+}$, while $\langle\tilde{R}^+\rangle_s = \langle\tilde{R}^-\rangle_s = 0$. Here, we took into account the relations: $\langle\tilde{R}^+\tilde{R}^-\rangle = \frac{1}{2}(1 + \tilde{R}_z)$, $\langle\tilde{R}^-\tilde{R}^+\rangle = \frac{1}{2}(1 - \tilde{R}_z)$. The time dependence of the mean-values of operators $\tilde{R}^\pm$ and $\tilde{R}_z$ in the "double dressed" state basis follows immediately from (7):

$$\begin{aligned} \langle\tilde{R}^+\rangle &= \langle\tilde{R}^+(0)\rangle e^{(2i\bar{G}_R - \bar{\Gamma}_s)\tau}, \\ \langle\tilde{R}^-\rangle &= \langle\tilde{R}^-(0)\rangle e^{-(2i\bar{G}_R + \bar{\Gamma}_s)\tau}, \\ \langle\tilde{R}_z\rangle &= \langle R_z(0)\rangle e^{-2(\bar{\Gamma}_- + \bar{\Gamma}_+)\tau} + \langle\tilde{R}_z\rangle_s\left(1 - e^{-2(\bar{\Gamma}_- + \bar{\Gamma}_+)\tau}\right), \end{aligned} \tag{8}$$

where $\bar{\Gamma}_s = 4\bar{\Gamma}_0 + \bar{\Gamma}_+ + \bar{\Gamma}_-$.

3. Resonance Fluorescence

The phenomenon of resonance fluorescence is the process in which a laser-pumped two-level atom scatters photons both coherently and incoherently. If the driving field is monochromatic, at low excitement intensities, the atom absorbs a photon and reemits it at that frequency as a consequence of energy conservation. The spectral width of the fluorescent light is very narrow. The situation is considerably more complex when the laser intensity increases and the Rabi frequency associated with the laser field is comparable or greater than the width of the energy band of the quantum emitter. In this case, Rabi oscillations appear as a modulation of the dipolar quantum moment and side bands appear in the spectrum of the emitted radiation. The resonance fluorescence spectrum is represented via the terms of the double-correlated function of the emitted field [12]:

$$S(\omega) = \Phi(r) Re \int_0^\infty d\,\tau e^{i(\omega-\omega_L)\tau} \lim_{t\to\infty} \langle S^+(t) S^-(t-\tau)\rangle, \tag{9}$$

where $\Phi(r) = \frac{2d^2\omega_{21}^4}{3r^2c^4}$, r is the distance to the detector. In the double-dressed picture, we found the following expression for the correlation function entering in (9):

$$\lim_{t\to\infty}\langle S^+(t)S^-(t)\rangle =$$
$$= \left(\frac{1}{4}\sin^2 2\,\theta\cos^2 2\bar{\theta} + \frac{1}{4}\sin^2 2\bar{\theta}\cos^4\theta e^{-i\,\omega\tau} + \frac{1}{4}\sin^2 2\bar{\theta}\sin^4\theta e^{i\,\omega\tau}\right)\langle \tilde{R}_z\tilde{R}_z(\tau)\rangle +$$
$$+\left(\frac{1}{4}\sin^2 2\,\theta\sin^2 2\bar{\theta} + \cos^4\theta\sin^4\bar{\theta}e^{-i\,\omega\tau} + \sin^4\theta\cos^4\bar{\theta}e^{i\,\omega\tau}\right)\langle \tilde{R}^-\tilde{R}^+(\tau)\rangle +$$
$$+\left(\frac{1}{4}\sin^2 2\bar{\theta}\sin^2 2\,\theta + \cos^4 4\bar{\theta}\cos^4\theta e^{-i\,\omega\tau} + \sin^4\theta\sin^4\bar{\theta}e^{i\,\omega\tau}\right)\langle \tilde{R}^+\tilde{R}^-(\tau)\rangle. \tag{10}$$

Using the time-dependence of the qubit operators in (10) and integrating the corresponding expressions, we obtain the final expression of resonance fluorescence spectra in the steady-state:

$$S(\nu) = \frac{1}{4}\sin^2 2\,\theta\cos^2 2\bar{\theta}\left\{\pi\langle\tilde{R}_z\rangle_s^2\delta(\nu-\omega_L) + \left(1-\langle\tilde{R}_z\rangle_s^2\right)\frac{\Gamma_\parallel}{\Gamma_\parallel^2+(\nu-\omega_L)^2}\right\} +$$
$$+\frac{1}{4}\sin^2 2\bar{\theta}\cos^4\theta\left\{\pi\langle\tilde{R}_z\rangle_s^2\delta(\nu-\omega_L-\omega) + \left(1-\langle\tilde{R}_z\rangle_s^2\right)\frac{\Gamma_\parallel}{\Gamma_\parallel^2+(\nu-\omega_L-\omega)^2}\right\} +$$
$$+\frac{1}{4}\sin^2 2\bar{\theta}\sin^4\theta\left\{\pi\langle\tilde{R}_z\rangle_s^2\delta(\nu-\omega_L+\omega) + \left(1-\langle\tilde{R}_z\rangle_s^2\right)\frac{\Gamma_\parallel}{\Gamma_\parallel^2+(\nu-\omega_L+\omega)^2}\right\} +$$
$$+\langle\tilde{R}^-\tilde{R}^+\rangle_s\left[\frac{1}{4}\sin^2 2\,\theta\sin^2 2\bar{\theta}\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega_L+2\bar{G}_R)^2} +\right.$$
$$+\cos^4\theta\sin^4\bar{\theta}\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega_L-\omega+2\bar{G}_R)^2} +$$
$$\left.+\sin^4\theta\cos^4\bar{\theta}\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega_L+\omega+2\bar{G}_R)^2}\right] +$$
$$+\langle\tilde{R}^+\tilde{R}^-\rangle_s\left[\frac{1}{4}\sin^2 2\bar{\theta}\sin^2 2\,\theta\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega_L-2\bar{G}_R)^2} +\right.$$
$$+\cos^4\bar{\theta}\cos^4\theta\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega_L-\omega-2\bar{G}_R)^2} +$$
$$\left.+\sin^4\theta\sin^4\bar{\theta}\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega_L+\omega-2\bar{G}_R)^2}\right], \tag{11}$$

note that: $\Gamma_\parallel = 2(\bar{\Gamma}_- + \bar{\Gamma}_+)$.

Analyzing the resonance fluorescence spectrum given in (11), one can observe that there are three coherently scattered spectral lines at ω_L, and $\omega_L \pm \omega$ nad up to nine incoherently scattered spectral bands, i.e., at $\nu-\omega_L = 0$, $\nu-\omega_L-\omega = 0$, $\nu-\omega_L+\omega = 0$, etc., in agreement with the double-dressed state picture. Particularly, Fig. 1 depicts the resonance fluorescence spectrum for certain parameters. More specifically, one can observe the cancelation of the central spectral band due to interference effects among the induced double dressed-state transitions. Asymmetrical behaviors in the scattered light spectrum are observed as well. This is because of the population inversion in the bare state and it differs from the usual resonance fluorescence spectrum obtained in the absence of permanent dipoles [2, 13].

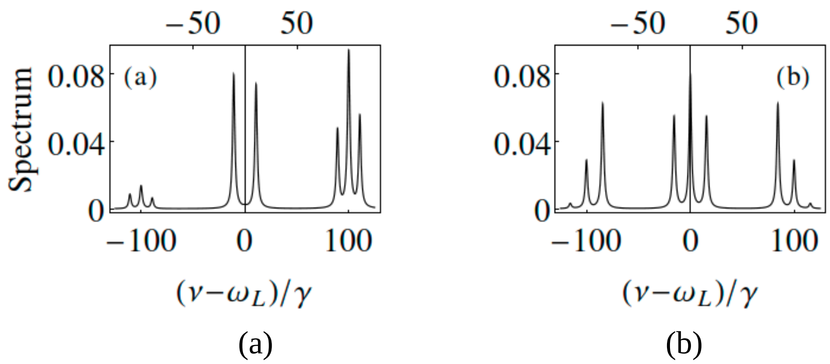

Fig. 1. Resonance fluorescence spectra. Here $\Omega/\gamma = 45$, $\omega/\gamma = 100$ and (a) $\Delta/(2\Omega) = 0{,}5$; (b) $\Delta/(2\Omega) = 0$

Finalizing this part, we have studied the resonance fluorescence properties of this system. In the following, we shall calculate the squeezing effects in the resonance fluorescence processes of laser-pumped molecules with permanent dipoles.

4. Squeezing Spectra and Quantum Fluctuations

The steady-state spectrum of squeezing can be determined as follows [14]:

$$S_\varphi(\nu) = (\gamma/|\mu|^2)\int_0^\infty [exp(i\nu\tau) + exp(-i\nu\tau)] \times \lim_{t\to\infty} \Gamma_\varphi(t+\tau,t)\, d\tau, \tag{12}$$

where $\Gamma_\varphi(t+\tau,t) = \langle : \Delta E_\varphi(t+\tau)\Delta E_\varphi(t) : \rangle$. Reduced quantum fluctuations are phase dependent due to the weak oscillating terms of the field emitted to the detector:

$$E_\varphi(t) = \frac{E^{(+)}(t)exp(i\,\varphi) + E^{(-)}(t)exp(-i\,\varphi)}{2}, \tag{13}$$

where φ is the phase angle and $E^{(\pm)}$ are the negative and positive amplitudes of the field. Particularly, one can show that

$$\Gamma_\varphi(t+\tau,t) = \frac{1}{4}\times \langle : \big(E^{(+)}(t+\tau)e^{i\varphi} - E^{(-)}(t+\tau)e^{-i\varphi} - \langle E^{(+)}(t+\tau)\rangle e^{i\varphi} - \langle E^{(-)}(t+\tau)\rangle e^{-i\varphi}\big) \times \big(E^{(+)}(t)e^{i\varphi} - E^{(-)}(t)e^{-i\varphi} - \langle E^{(+)}(t)\rangle e^{i\varphi} - \langle E^{(-)}(t)\rangle e^{-i\varphi}\big) : \rangle, \tag{14}$$

or,

$$\Gamma_\varphi(t+\tau,t) = \frac{1}{4}\times \big\{e^{2i\varphi}\langle E^{(+)}(t+\tau)E^{(+)}(t)\rangle + e^{-2i\varphi}\langle E^{(-)}(t+\tau)E^{(-)}(t)\rangle + +\langle E^{(-)}(t+\tau)E^{(+)}(t)\rangle + \langle E^{(-)}(t)E^{(+)}(t+\tau)\rangle\big\}. \tag{15}$$

In the strong field approximation and in the absence of the free field, the radiated field $E^{(+)}(t)$ can be substituted by $\mu S_{-}(t)$, where μ is a geometric factor. Since $E^{+} \sim a \sim S^{-}$; $E \sim a^{+} \sim S^{+}$, in this way, we obtain:

$$\begin{aligned}
&\Gamma_{\varphi}(t+\tau,t) = \\
&\frac{|\mu|^2}{4} \times \{e^{2i\varphi}(\langle S^{-}(t+\tau)S^{-}(t)\rangle - \langle S^{-}(t+\tau)\rangle\langle S^{-}(t)\rangle) + \\
&+e^{-2i\varphi}(\langle S^{+}(t)S^{+}(t+\tau)\rangle - \langle S^{+}(t)\rangle\langle S^{+}(t+\tau)\rangle) + \\
&+(\langle S^{+}(t+\tau)S^{-}(t)\rangle - \langle S^{+}(t+\tau)\rangle\langle S^{-}(t)\rangle) + \\
&+(\langle S^{+}(t)S^{-}(t+\tau)\rangle - \langle S^{+}(t)\rangle\langle S^{-}(t+\tau)\rangle)\}.
\end{aligned} \tag{16}$$

Analytical expression for the spectrum of squeezing is computed according to (12)–(16) using the solutions of the equations of motion (8) [15]. Finally, we obtain

$$\begin{aligned}
S_{\varphi}(\nu) = \frac{\gamma}{4} \times \{&(\langle \tilde{R}_z^2\rangle_s - \langle \tilde{R}_z\rangle_s^2)[\sin^2 2\theta \cos^2 2\bar{\theta}\,(1+\cos 2\varphi)\chi_1(\nu) + \\
&+\frac{1}{2}\sin^2 2\bar{\theta}\left(\cos^4\theta + \sin^4\theta + \frac{1}{2}\sin^2 2\theta\cos 2\varphi\right)\chi_2(\nu) + \\
&+\frac{1}{2}\sin^2 2\theta \sin^2 2\bar{\theta}\,(1+\cos 2\varphi)\chi_3(\nu) + \\
&+\sin^4\bar{\theta}\left(\cos^4\theta + \sin^4\theta - \langle \tilde{R}_z\rangle_s \cos 2\theta - \frac{1}{2}\sin^2 2\theta\cos 2\varphi\right)\chi_4(\nu) + \\
&+\cos^4\bar{\theta}\left(\cos^4\theta + \sin^4\theta + \langle \tilde{R}_z\rangle_s \cos 2\theta - \frac{1}{2}\sin^2 2\theta\cos 2\varphi\right)\chi_5(\nu) + \\
&+\frac{1}{2}\sin^2 2\theta\sin^2 2\bar{\theta}\sin 2\varphi\,\langle \tilde{R}_z\rangle_s\chi_6(\nu) + \frac{1}{2}\sin^2 2\theta\sin^4\bar{\theta}\sin 2\varphi\,\langle \tilde{R}_z\rangle_s\chi_7(\nu) + \\
&+\frac{1}{2}\sin^2 2\theta\cos^4\bar{\theta}\sin 2\varphi\,\langle \tilde{R}_z\rangle_s\chi_8(\nu),
\end{aligned} \tag{17}$$

where

$\chi_1(\nu) = \frac{2\bar{\Gamma}_{\parallel}}{\bar{\Gamma}_{\parallel}^2+\nu^2}$, $\chi_2(\nu) = \frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega)^2} + \frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu+\omega)^2}$, $\chi_3(\nu) = \frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-2\bar{G}_R)^2} + \frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu+2\bar{G}_R)^2}$, $\chi_4(\nu) = \frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega+2\bar{G}_R)^2} + \frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu+\omega-2\bar{G}_R)^2}$, $\chi_5(\nu) = \frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu+\omega+2\bar{G}_R)^2} + \frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega-2\bar{G}_R)^2}$, $\chi_6(\nu) = \frac{(\nu-2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu-2\bar{G}_R)^2} - \frac{(\nu+2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu+2\bar{G}_R)^2}$, $\chi_7(\nu) = \frac{(\nu-\omega+2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu-\omega+2\bar{G}_R)^2} - \frac{(\nu+\omega-2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu+\omega-2\bar{G}_R)^2}$, $\chi_8(\nu) = \frac{(\nu+\omega+2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu+\omega+2\bar{G}_R)^2} - \frac{(\nu-\omega-2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu-\omega-2\bar{G}_R)^2}$, $\langle \tilde{R}_z\rangle_s = \frac{\bar{\Gamma}_- - \bar{\Gamma}_+}{\bar{\Gamma}_- + \bar{\Gamma}_+}$.

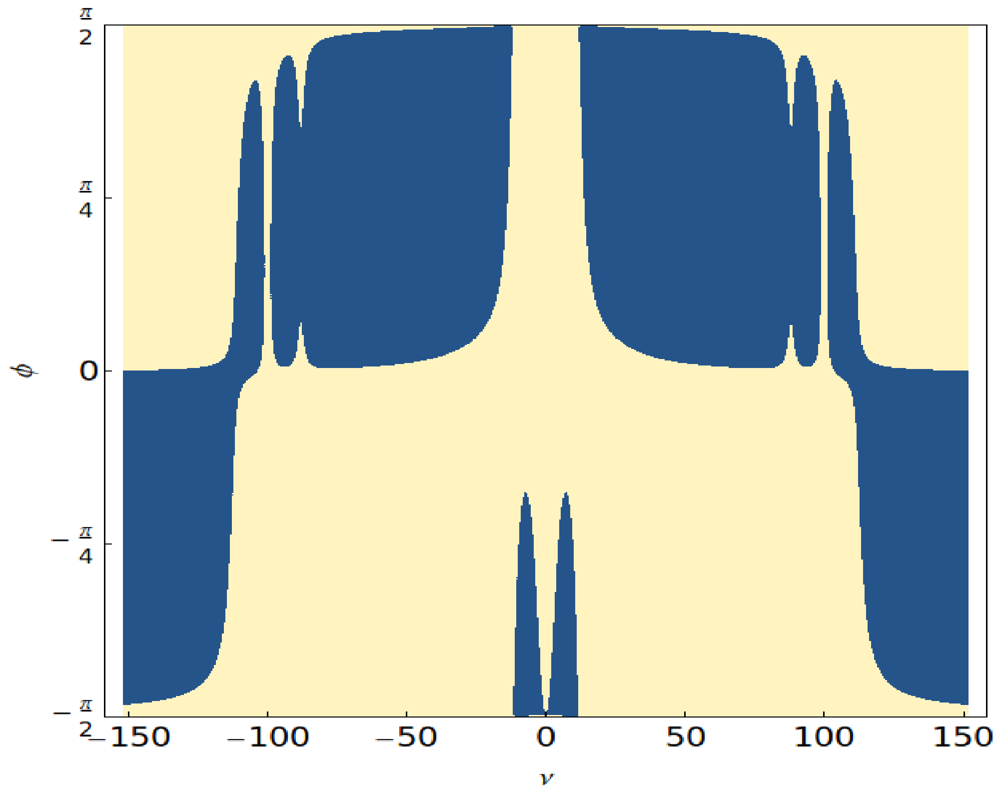

Fig. 2. Spectrum of squeezing in resonance fluorescence processes with permanent dipole systems. Parameters of interest are $\Omega/\gamma = 45$, $\omega/\gamma = 100$, $q = 0{,}7$.

In Fig. 2, we plot the squeezing spectrum for certain parameters of interest. Particularly, squeezing occurs for negative values (dark area in Fig. 2) and broader squeezing ranges take place because of permanent dipoles (see also [15]). Especially, squeezing around ν is due to permanent dipoles and will not be observed in the absence of it.

On the other hand, the total quantum fluctuation expression can be obtained after frequency integrating of (17), i.e.,

$$
\begin{gathered}
\langle :\left(\Delta E_{\varphi}\right)^{2}: \rangle = \pi \frac{d^2}{2\pi\gamma} \times \\
\{\left(\langle \tilde{R}_z^2 \rangle_s - \langle \tilde{R}_z \rangle_s^2\right)[4 \sin^2 2\theta \cos^2 2\bar{\theta} \cos^2 \varphi + \\
+ \sin^2 2\bar{\theta} \left(\cos^4 \theta + \sin^4 \theta + \frac{1}{2} \sin^2 2\theta \cos 2\varphi\right) + \sin^2 2\theta \sin^2 2\bar{\theta} \left(1 + \cos 2\varphi\right) + \\
+ 2 \sin^4 \bar{\theta} \left(\cos^4 \theta + \sin^4 \theta - \langle \tilde{R}_z \rangle_s \cos 2\theta - \frac{1}{2} \sin^2 2\theta \cos 2\varphi\right) + \\
+ 2 \cos^4 \bar{\theta} \left(\cos^4 \theta + \sin^4 \theta + \langle \tilde{R}_z \rangle_s \cos 2\theta - \frac{1}{2} \sin^2 2\theta \cos 2\varphi\right) + \\
+ \sin^2 2\theta \sin^2 2\bar{\theta} \sin 2\varphi \langle \tilde{R}_z \rangle_s + \sin^2 2\theta \sin^4 \bar{\theta} \sin 2\varphi \langle \tilde{R}_z \rangle_s + \\
+ \sin^2 2\theta \cos^4 \bar{\theta} \sin 2\varphi \langle \tilde{R}_z \rangle_s .
\end{gathered} \tag{18}
$$

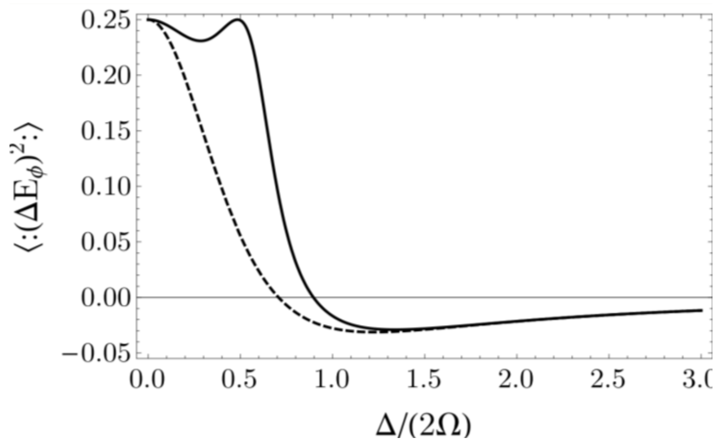

Fig. 3. The variance $\langle:\left(\Delta \mathrm{E}_{\phi}\right)^{2}:\rangle$ in units of $|d|^2$ as a function of $\Delta/2\Omega$ for $G/\gamma = 0$ (dashed line) $G/\gamma = 16$ (solid line). Other parameters are $\varphi = -\frac{\pi}{4}$, $\omega/\gamma = 100$, $q = 0.7$, $\Omega/\gamma = 45$.

In Fig. 3, we depict variance $\langle:\left(\Delta \mathrm{E}_{\phi}\right)^{2}:\rangle$ proceeding from the resonance fluorescence processes of laser-pumped two-level systems possessing permanent dipoles. We have found distinct quantum fluctuations features, which are due to permanent dipoles (compare the solid and dashed curves in Fig. 3, see also [16]).

5. Summary

We have studied the resonanse fluorescence spectrum of spontaenously emitted photons during the laser pumping processes of two-level molecules with permanent dipoles. Features differing from those in the case of similar processes yet in the absence of permanent dipoles have been found. In particular, additional spectral lines are emitted and extra squeezed frequency domains are observed.

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