

Effect of the exciton-exciton interaction on resonance fluorescence of excitons in a quantum well

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The effect of the exciton-exciton interaction on the resonance fluorescence of quantum-well excitons is studied. At relative low exciton density, the resonance fluorescence exists only when the exciton-exciton interaction is not negligible. The fluorescence spectrum changes from one peak to two peaks when the pump light intensity is increased beyond a critical value. The fluorescence light is antibunching when the mean exciton number in the quantum well is more than 0.5. The difference between exciton resonance fluorescence and two-level atom resonance fluorescence is presented.

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I. INTRODUCTION

It is well known that an excited two-level atom will spontaneously radiate and fall to its ground state eventually. When a two-level atom is driven by a resonant electromagnetic field, it will radiate continuously and reach a steady state. In 1969, Mollow predicted that the fluorescence spectrum of a two-level atom driven by a strong coherent field consists of three peaks, when the corresponding Rabi frequency is larger than the spontaneous emission rate [1]. This prediction had been verified by many experiments [2–4]. Since then, a great deal of research work has been done on the resonance fluorescence of atoms or molecules [5–12].

In recent years, the optical properties of excitons have become a subject of extensive investigation because of the potential technological impact of the excitonic devices on optoelectronics and photonics. Especially, people have studied the optical properties of excitons in confined structures such as quantum well, quantum wire, or quantum dot.

The radiation of the exciton exhibits the superradiance character. The initial studies were mainly focused on the superradiance of Wannier excitons in semiconductor microcrystallites, both theoretically [13] and experimentally [14]. Then, the superradiance of Frenkel excitons was observed in J aggregates at low temperature [15,16]. However, higher or complete population inversion may make this system unstable [17]. In 1995, the superradiance of high-density Frenkel excitons in an R -phycoerythrin (R -PE) single crystal was observed at room temperature for the first time [18].

Tokihiko *et al.* [17] have studied theoretically the superradiance behavior of the Frenkel exciton in a linear meso-

scopic chain. They first investigated the exciton superradiance at low density where exciton-exciton interaction is negligible [17]. Then, at higher exciton density, they took into account the interaction between Frenkel excitons [19]. The emission of Frenkel excitons in a one-dimensional mesoscopic system interacting via the static dipole moments [20] was also studied.

Excitons confined in a thin layer of thickness $L < \lambda$ have also been studied [21]. In the case of the Frenkel exciton, the radiative decay rate is on the order of $\Gamma \sim (\lambda^2/a^2)\gamma_0$, where a is the lattice constant and γ_0 is the decay rate of an isolated atom or molecule. Knoester has studied the dependence of the superradiance rate on the layer thickness L [22]. He found that the superradiance rate first increases with L , and then drops. Through damped oscillation, it eventually vanishes when $L \rightarrow \infty$, which is consistent with the well-known result that the exciton and the emitted photon will mix to form stable polaritons.

In this paper we will study the effect of exciton-exciton interaction on the resonance fluorescence of the Wannier excitons in the quantum well. In Sec. II the general formulation is presented. The excitons are described by bosonic operators in the low-density limit [23]. When the exciton density increases, excitons no longer behave like ideal bosonic particles. One way to deal with this deviation is to introduce an effective interaction between the hypothetical ideal bosons [24,25]. In this work we take this approach to derive the master equation for the reduced density operators of the excitons. Then, we convert the operator equation into a Fokker-Planck equation via the positive P representation. In Sec. III the stochastic differential equations corresponding to the Fokker-Planck equation are proposed. We then linearize the stochastic differential equations near their steady-state solutions. In Sec. IV, we calculate the spectrum of the emitted light with these linear equations. We find no Rabi splitting in the emission spectrum in the low exciton density region.

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However, the interaction between excitons will induce the splitting of the emission spectrum to two peaks when the pumping field intensity exceeds a critical value. Finally the quantum intensity fluctuation properties of the steady fluorescence is discussed.

II. HAMILTONIAN AND FOKKER-PLANCK EQUATION

We assume the exciton density is not too high so that the deviation of excitons from ideal bosons can be treated by introducing an effective interaction between the excitons [24,25]. Under the rotating-wave approximation, the Hamiltonian of the system in which the excitons are driven by a monochromatic coherent light is written as :

$$\begin{aligned} \hat{H} = & \hbar\Omega\hat{b}^+\hat{b} + \hbar G\hat{b}^+\hat{b}^+\hat{b}\hat{b} + i\hbar(E\hat{b}^+e^{-i\Omega t} - E^*\hat{b}e^{i\Omega t}) \\ & + \hbar\sum_k k\omega_k\hat{a}_k^+\hat{a}_k + \hbar\hat{b}^+\sum_k g_k\hat{a}_k + \hbar\hat{b}\sum_k g_k^*\hat{a}_k^+, \end{aligned} \quad (1)$$

\hat{b}^+ (\hat{b}) are creation (annihilation) operators for the excitons. They are assumed to obey the bosonic commutation relation, $[\hat{b}, \hat{b}^+] = 1$. \hat{a}_k^+ (\hat{a}_k) are the creation (annihilation) operators for the photons in mode k , propagating perpendicular to the quantum-well plane under the condition that excitons have no momentum parallel to the quantum-well plane. G represents the interaction constant between the excitons, including the residual Coulomb interaction. E is proportional to the amplitude of the driving field. g_k is the coupling constant between the photons and the excitons.

The time evolution of the exciton system is described by this master equation of the reduced density operator. In the Schrödinger picture this master equation, which is based on the Born-Markoff approximation, is written as

$$\frac{\partial \hat{\rho}(t)}{\partial t} = \frac{1}{i\hbar} [\hat{H}_0, \hat{\rho}] + \gamma(2\hat{b}\hat{\rho}\hat{b}^+ - \hat{\rho}\hat{b}^+\hat{b} - \hat{b}^+\hat{b}\hat{\rho}), \quad (2)$$

where

$$\begin{aligned} \hat{H}_0 = & \hbar\Omega\hat{b}^+\hat{b} + \hbar G\hat{b}^+\hat{b}^+\hat{b}\hat{b} \\ & + i\hbar(E\hat{b}^+e^{-i\Omega t} - E^*\hat{b}e^{i\Omega t}), \end{aligned}$$

γ is the spontaneous decay rate of the exciton. Since the exciton is boson, its density operator can be represented by the c -number function like the photon density operator. So we may convert Eq. (2) into a Fokker-Planck equation of the function $P(\alpha, \alpha^+, t)$ by use of the positive P presentation [26],

$$\begin{aligned} \frac{\partial P(\alpha, \alpha^+, t)}{\partial t} = & \left\{ \frac{\partial}{\partial \alpha} (\gamma\alpha - E + i2G\alpha^2\alpha^+) \right. \\ & + \frac{\partial}{\partial \alpha^+} (\gamma\alpha^+ - E^* - i2G\alpha\alpha^+) \\ & \left. - \frac{\partial^2}{\partial \alpha^2} (iG\alpha^2) + \frac{\partial^2}{\partial \alpha^+{}^2} (iG\alpha\alpha^+) \right\} \end{aligned}$$

$$\times P(\alpha, \alpha^+, t), \quad (3)$$

where α corresponds to \hat{b} , and α^+ corresponds to \hat{b}^+ . α and α^+ are considered as independent complex variables. We have also transformed into the rotating frame of the exciton variables with $\alpha \rightarrow \alpha e^{-i\Omega t}$ and $\alpha^+ \rightarrow \alpha^+ e^{i\Omega t}$.

III. STOCHASTIC DIFFERENTIAL EQUATIONS AND ANALYSIS OF THE FLUCTUATION

The nonlinear Fokker-Planck equation can be solved exactly only in some special cases. It is difficult to obtain an analytical expression of $P(\alpha, \alpha^+, t)$ from the Fokker-Planck equation (3). Hence, we will make an approximation. We first use the Ito rules to convert this Fokker-Planck equation into stochastic differential equations. Such conversion is allowed because the Fokker-Planck equation in the positive P representation always has a positive semidefinite diffusion matrix. The obtained stochastic differential equations for α and α^+ are

$$\begin{aligned} \frac{\partial}{\partial t} \begin{pmatrix} \alpha \\ \alpha^+ \end{pmatrix} = & \begin{pmatrix} -\gamma\alpha + E - i2G\alpha^2\alpha^+ \\ -\gamma\alpha^+ + E^* + i2G\alpha\alpha^+ \end{pmatrix} \\ & + \begin{pmatrix} -i2G\alpha^2 & 0 \\ 0 & i2G\alpha\alpha^+ \end{pmatrix}^{1/2} \begin{pmatrix} \xi \\ \xi^+ \end{pmatrix}, \end{aligned} \quad (4)$$

where $\xi(t)$ and $\xi^+(t)$ are the Gaussian fluctuation forces, satisfying the following correlation functions:

$$\begin{aligned} \langle \xi(t) \rangle = \langle \xi^+(t) \rangle & = 0, \\ \langle \xi(t)\xi^+(t') \rangle = \langle \xi^+(t)\xi(t') \rangle & = 0, \\ \langle \xi(t)\xi(t') \rangle = \langle \xi^+(t)\xi^+(t') \rangle & = \delta(t-t'). \end{aligned} \quad (5)$$

Next, we discard the fluctuation forces to obtain the steady-state solution. In the case where the steady state is stable, the fluctuation forces will only cause fluctuations near this solution.

We denote the steady values of α and α^+ by α_0 and α_0^+ , respectively. From Eq. (4), it is evident that α_0 and α_0^+ are given by

$$\begin{aligned} E - \gamma\alpha_0 - i2G\alpha_0^2\alpha_0^+ & = 0, \\ E^* - \gamma\alpha_0^+ + i2G\alpha_0\alpha_0^+ & = 0. \end{aligned} \quad (6)$$

It is not difficult to see that α_0 and α_0^+ are complex conjugates of each other.

Now, we will proceed by making the small fluctuation approximation. When the system reaches the ‘‘steady’’ state, $\alpha(t)$ can be divided into two parts:

$$\begin{aligned} \alpha(t) & = \alpha_0 + \delta\alpha(t), \\ \alpha^+(t) & = \alpha_0^+ + \delta\alpha^+(t), \end{aligned} \quad (7)$$

where both $\delta\alpha(t)$ and $\delta\alpha^+(t)$ are fluctuations with zero mean values. Assuming the fluctuations are small, we obtain the linearized equation for $\delta\alpha$ and $\delta\alpha^+$ from Eq. (4),

$$\begin{aligned} \frac{\partial}{\partial t} \begin{pmatrix} \delta\alpha(t) \\ \delta\alpha^+(t) \end{pmatrix} = - \begin{pmatrix} \gamma + i4G\alpha_0\alpha_0^+ & i2G\alpha_0^2 \\ -i2G\alpha_0^{+2} & \gamma - i4G\alpha_0^+\alpha_0 \end{pmatrix} \\ \times \begin{pmatrix} \delta\alpha(t) \\ \delta\alpha^+(t) \end{pmatrix} + \begin{pmatrix} -i2G\alpha_0^2 & 0 \\ 0 & i2G\alpha_0^{+2} \end{pmatrix}^{1/2} \\ \times \begin{pmatrix} \xi(t) \\ \xi^+(t) \end{pmatrix}. \end{aligned} \quad (8)$$

Before solving this equation, let us calculate the $n \equiv \alpha_0^+\alpha_0$, which is the so-called coherent part of the total exciton number. Since α_0 and α_0^+ are complex conjugates of each other in the steady state, n is a positive real number. From Eq. (6), we get

$$n^3 + \frac{\gamma^2}{4G^2}n - \frac{|E|^2}{4G^2} = 0. \quad (9)$$

There is only one root of Eq. (9) to be a positive real number:

$$\begin{aligned} n = \sqrt[3]{\frac{|E|^2}{8G^2} + \sqrt{\frac{|E|^4}{64G^4} + \left(\frac{\gamma^2}{12G^2}\right)^3}} \\ - \sqrt[3]{-\frac{|E|^2}{8G^2} + \sqrt{\frac{|E|^4}{64G^4} + \left(\frac{\gamma^2}{12G^2}\right)^3}}. \end{aligned} \quad (10)$$

Under weak field $|E|^2 \ll \gamma^3/(3\sqrt{3}G)$, n is approximately expressed by $|E|^2/\gamma^2$, which is independent of G and increases with $|E|^2$. Under strong field $|E|^2 \gg \gamma^3/(3\sqrt{3}G)$, n is approximately expressed by $(|E|/2G)^{2/3}$, which is inversely proportional to $G^{2/3}$. This indicates a suppression of the coherent exciton number by the exciton-exciton interaction.

Now, we discuss the stability of the system. The stability means when α and α^+ deviate from α_0 and α_0^+ , they will return to α_0 and α_0^+ in case the fluctuation term is dropped. Equation (8) with the fluctuation terms deleted is written as

$$\frac{\partial}{\partial t} \begin{pmatrix} \delta\alpha \\ \delta\alpha^+ \end{pmatrix} = - \begin{pmatrix} \gamma + i4Gn & i2G\alpha_0^2 \\ -i2G\alpha_0^{+2} & \gamma - i4Gn \end{pmatrix} \begin{pmatrix} \delta\alpha \\ \delta\alpha^+ \end{pmatrix}, \quad (11)$$

α_0 is expressed in terms of n by $E/(\gamma + i2Gn)$. The eigen-solutions of Eq. (11) are of the form $\delta\alpha(0)e^{-\Gamma t}$ and $\delta\alpha^+(0)e^{-\Gamma t}$. The eigenvalues of Γ are determined by

$$\begin{vmatrix} \gamma + i4Gn - \Gamma & i2G\alpha_0^2 \\ -i2G\alpha_0^{+2} & \gamma - i4Gn - \Gamma \end{vmatrix} = 0. \quad (12)$$

Only if all roots of Eq. (12) have the positive real parts, α_0 and α_0^+ are steady solutions. Equation (12) is a second-order equation of Γ . Its two roots are easily derived,

$$\Gamma_{1,2} = \gamma \pm i2\sqrt{3}Gn. \quad (13)$$

These results show that α_0 and α_0^+ are indeed the stable steady-state solutions. In the following two sections, we will discuss the resonance fluorescence of the excitons and the fluctuation of the light intensity.

IV. RESONANCE FLUORESCENCE OF EXCITONS

The radiated field operator at position z outside the quantum well is related to the exciton operator by the retardation relation [27,28]

$$\hat{E}_R^{(+)}(z,t) = \beta(z)\hat{b}\left(t - \frac{z}{c}\right), \quad (14)$$

where the index (+) of $\hat{E}_R^{(+)}$ indicates the positive frequency part, z represents the coordinate perpendicular to the quantum-well plane, and $\beta(z)$ is a parameter independent of t . The frequency spectrum of the steady resonance fluorescence is defined as the Fourier transformation of the two-time correlation function $\langle \hat{E}_R^{(-)}(z,t_0 + \tau)\hat{E}_R^{(+)}(z,t_0) \rangle$ in the limit $t_0 \rightarrow \infty$,

$$\begin{aligned} I_R(z,\omega) = \frac{1}{2\pi} \lim_{t_0 \rightarrow \infty} \\ \times \int_{-\infty}^{+\infty} \langle \hat{E}_R^{(-)}(z,t_0 + \tau)\hat{E}_R^{(+)}(z,t_0) \rangle e^{-i\omega\tau} d\tau. \end{aligned} \quad (15)$$

Substituting Eq. (14) into Eq. (15) yields

$$I_R(\omega) = \lim_{t_0 \rightarrow \infty} \frac{\beta^2}{2\pi} \int_{-\infty}^{+\infty} \langle \hat{b}^+(t_0 + \tau)\hat{b}(t_0) \rangle e^{-i\omega\tau} d\tau. \quad (16)$$

The integral in Eq. (16) can be expressed in terms of the positive P representation :

$$\begin{aligned} \lim_{t_0 \rightarrow \infty} \int_{-\infty}^{+\infty} \langle b^+(t_0 + \tau)b(t_0) \rangle e^{-i\omega\tau} d\tau \\ = 2\pi n \delta(\omega) + \int_{-\infty}^{+\infty} \langle \delta\alpha^+(\tau)\delta\alpha(0) \rangle e^{-i\omega\tau} d\tau. \end{aligned} \quad (17)$$

The first term of Eq. (17) represents the elastic ‘‘scattering’’ spectrum, thus it should be omitted from the fluorescence spectrum. To calculate $\int_{-\infty}^{+\infty} \langle \delta\alpha^+(\tau)\delta\alpha(0) \rangle e^{-i\omega\tau} d\tau$, Eq. (8) is written as follows:

$$\frac{\partial}{\partial t} \begin{pmatrix} \delta\alpha \\ \delta\alpha^+ \end{pmatrix} = -A \begin{pmatrix} \delta\alpha \\ \delta\alpha^+ \end{pmatrix} + D^{1/2} \begin{pmatrix} \xi \\ \xi^+ \end{pmatrix}, \quad (18)$$

where

$$A = \begin{bmatrix} \gamma + i4Gn & i2G\alpha_0^2 \\ -i2G\alpha_0^{+2} & \gamma - i4Gn \end{bmatrix},$$

and

$$D = \begin{bmatrix} -i2G\alpha_0^2 & 0 \\ 0 & i2G\alpha_0^{+2} \end{bmatrix}.$$

The matrix of the spectrum function of the radiation-field fluctuation is defined as

$$S(\omega) = \frac{1}{|\lambda|^2} \begin{bmatrix} -i2G\alpha_0^2[(\gamma - i4Gn)^2 + \omega^2 + 4G^2n^2] & 8G^2n^2\gamma \\ 8G^2n^2\gamma & i2G\alpha_0^{+2}[(\gamma + i4Gn)^2 + \omega^2 + 4G^2n^2] \end{bmatrix}, \quad (21)$$

where $|\lambda|^2 = (\gamma^2 - \omega^2 + 12G^2n^2)^2 + 4\gamma^2\omega^2$. The second term of Eq. (17) is given by the nondiagonal matrix element $S_{21}(\omega)$ of Eq. (19). Thus after transforming back to the original frame, the fluorescence spectrum of the exciton is

$$I_0(\omega) = \frac{\beta^2}{2\pi} \frac{8G^2n^2\gamma}{[\gamma^2 - (\omega - \Omega)^2 + 12G^2n^2]^2 + 4\gamma^2(\omega - \Omega)^2}. \quad (22)$$

The peak of the $I_0(\omega)$ corresponds to the minimum value of the denominator in Eq. (22). Under weak pump field ($12G^2n^2 \leq \gamma^2$), the fluorescence spectrum consists of a single peak centered at $\omega = \Omega$. Under strong pump field ($12G^2n^2 > \gamma^2$), the fluorescence spectrum has two peaks located at

$$\omega = \Omega \pm \sqrt{12G^2n^2 - \gamma^2}. \quad (23)$$

The curves $I_0(\omega) - \omega$ are shown in Fig. 1. We note that there are no Rabi splitting peaks in $I_0(\omega)$ despite the fact that the energy levels of the interacting photon-exciton system have in Rabi splitting. The absence of Rabi splitting peaks in $I_0(\omega)$ is due to the bosonic nature of excitons, rather than our small fluctuation approximation. To illustrate this point, we consider a simple case $G=0$; namely, the exciton-exciton interaction can be neglected. In this case, the exact solution of field operators can be obtained. The system Hamiltonian now becomes

$$\hat{H}_0 = \hbar\Omega\hat{b}^+\hat{b} + \hbar(E\hat{b}^+e^{-i\Omega t} - E^*\hat{b}e^{i\Omega t}). \quad (24)$$

To compare with the usual resonance fluorescence of a two-level atom, we will derive the first-order correlation function by the Langevin formulation. Since \hat{b} and \hat{b}^+ obey the bosonic commutation relation

$$S(\omega) \equiv \int_{-\infty}^{\infty} \begin{pmatrix} \langle \delta\alpha(\tau)\delta\alpha(0) \rangle & \langle \delta\alpha(\tau)\delta\alpha^+(0) \rangle \\ \langle \delta\alpha^+(\tau)\delta\alpha(0) \rangle & \langle \delta\alpha^+(\tau)\delta\alpha^+(0) \rangle \end{pmatrix} \times e^{-i\omega\tau} d\tau. \quad (19)$$

$S(\omega)$ will be given in terms of D and A by [29]

$$S(\omega) = (A + i\omega I)^{-1} D (A^T - i\omega I)^{-1}, \quad (20)$$

where I is the identity matrix and T means transpose. We get accordingly

$$[\hat{b}(t), \hat{b}^+(t)] = 1, \quad (25)$$

the Langevin equations in rotating frame, which corresponds to Eq. (24), are

$$\frac{d}{dt}\hat{b}^+(t) = -\gamma\hat{b}^+(t) + iE^* + \hat{F}^+(t), \quad (26)$$

and its Hermitian conjugate. $\hat{F}^+(t)$ is the Langevin fluctuation force.

The exact solution for $\langle \hat{b} \rangle$ is

$$\langle \hat{b}^+(t_0 + \tau) \rangle = \langle \hat{b}^+(t_0) \rangle e^{-\gamma\tau} - \frac{iE^*}{\gamma} (e^{-\gamma\tau} - 1). \quad (27)$$

The first-order correlation function is then derived by the quantum regression theorem:

$$\langle \hat{b}^+(t_0 + \tau)\hat{b}(t_0) \rangle = e^{-\gamma\tau} \langle \hat{b}^+(t_0)\hat{b}(t_0) \rangle - \frac{iE^*}{\gamma} (e^{-\gamma\tau} - 1) \langle \hat{b}(t_0) \rangle. \quad (28)$$

It is straightforward to obtain the equation for $\langle \hat{b}^+(t)\hat{b}(t) \rangle$ from Eq. (25). In the case that the reservoir is the vacuum ($n_T=0$), we get

$$\lim_{t_0 \rightarrow \infty} \langle \hat{b}^+(t_0)\hat{b}(t_0) \rangle = \frac{|E|^2}{\gamma^2}. \quad (29)$$

Thus, changing back to the original frame, we have

$$\lim_{t_0 \rightarrow \infty} \langle \hat{b}^+(t_0 + \tau)\hat{b}(t_0) \rangle = \frac{|E|^2}{\gamma^2} e^{i\Omega\tau}. \quad (30)$$

Substituting the above equation into Eq. (16) leads to

$$I_R(\omega) = \beta^2 \frac{|E|^2}{\gamma^2} \delta(\omega - \Omega). \quad (31)$$

We see there is no resonance fluorescence spectrum, let alone the Rabi splitting of the spectral peak.

The corresponding \hat{H}_0 for a two-level atom is similar to Eq. (24), the only difference is \hat{b}^+ and \hat{b} being replaced by the atomic level change operators $\hat{\sigma}_+$ and $\hat{\sigma}_-$. The difference between $\hat{\sigma}_+$, $\hat{\sigma}_-$ and \hat{b}^+ , \hat{b} lies in their commutation relations. For two-level atoms,

$$\begin{aligned} [\hat{\sigma}_-(t), \hat{\sigma}_+(t)] &= -\hat{\sigma}_3(t), \\ [\hat{\sigma}_3(t), \hat{\sigma}_\pm(t)] &= \pm 2\hat{\sigma}_\pm(t) \end{aligned} \quad (32)$$

The above Fermionic commutation relations results in coupled equations for $\hat{\sigma}_+$, $\hat{\sigma}_-$, and $\hat{\sigma}_3$:

$$\frac{d}{dt} \langle \hat{\sigma}_+(t) \rangle = -iE^* \langle \hat{\sigma}_3(t) \rangle - \gamma \langle \hat{\sigma}_+(t) \rangle, \quad (33)$$

$$\frac{d}{dt} \langle \hat{\sigma}_-(t) \rangle = iE \langle \hat{\sigma}_3(t) \rangle - \gamma \langle \hat{\sigma}_-(t) \rangle,$$

$$\frac{d}{dt} \langle \hat{\sigma}_3(t) \rangle = 2i[E^* \langle \hat{\sigma}_-(t) \rangle - E \langle \hat{\sigma}_+(t) \rangle] - 2\gamma \langle \hat{\sigma}_3(t) \rangle - 2\gamma.$$

It is just these coupled linear equations that yield three eigen-decay rates,

$$\gamma_1 = \gamma, \quad \Gamma_{2,3} = \frac{3}{2} \gamma \pm \frac{1}{2} \sqrt{\gamma^2 - 16|E|^2}. \quad (34)$$

When $|E|^2$ becomes larger than $\gamma^2/16$, Γ_2 and Γ_3 develop imaginary parts $\pm i\omega_R$, respectively, with $\omega_R = \sqrt{4|E|^2 - \gamma^2}/4$.

Corresponding to Eq. (27), for two-level atoms we get the solution of $\langle \hat{\sigma}_+(t) \rangle$ in the case $|E|^2 > \gamma^2/16$,

$$\begin{aligned} \langle \hat{\sigma}_+(t_0 + \tau) \rangle &= \frac{iE^* \gamma}{2|E|^2 + \gamma^2} \left\{ 1 - \frac{1}{2} e^{-(3/2)\gamma\tau} \left[\left(1 - \frac{3i\gamma}{2\omega_R} \right) e^{i\omega_R\tau} + \left(1 + \frac{3i\gamma}{2\omega_R} \right) e^{-i\omega_R\tau} \right] \right\} \\ &+ \left\{ \frac{1}{2} e^{-\gamma\tau} + \frac{1}{4} e^{-(3/2)\gamma\tau} \left[\left(1 - \frac{i\gamma}{2\omega_R} \right) e^{i\omega_R\tau} + \left(1 + \frac{i\gamma}{2\omega_R} \right) e^{-i\omega_R\tau} \right] \right\} \langle \hat{\sigma}_+(t_0) \rangle \\ &+ \frac{E^*}{E} \left\{ \frac{1}{2} e^{-\gamma\tau} - \frac{1}{4} e^{-(3/2)\gamma\tau} \left[\left(1 - \frac{i\gamma}{2\omega_R} \right) e^{i\omega_R\tau} + \left(1 + \frac{i\gamma}{2\omega_R} \right) e^{-i\omega_R\tau} \right] \right\} \langle \hat{\sigma}_-(t_0) \rangle \\ &- \frac{E^*}{2\omega_R} e^{-(3/2)\gamma\tau} (e^{i\omega_R\tau} - e^{-i\omega_R\tau}) \langle \hat{\sigma}_3(t_0) \rangle. \end{aligned} \quad (35)$$

By using the quantum regression theorem it turns out that

$$\begin{aligned} \langle \hat{\sigma}_+(t_0 + \tau) \hat{\sigma}_-(t_0) \rangle &= \frac{iE^* \gamma}{2|E|^2 + \gamma^2} \left\{ 1 - \frac{1}{2} e^{-(3/2)\gamma\tau} \left[\left(1 - \frac{3i\gamma}{2\omega_R} \right) e^{i\omega_R\tau} + \left(1 + \frac{3i\gamma}{2\omega_R} \right) e^{-i\omega_R\tau} \right] \right\} \langle \hat{\sigma}_-(t_0) \rangle \\ &+ \frac{1}{2} \left\{ \frac{1}{2} e^{-\gamma\tau} + \frac{1}{4} e^{-(3/2)\gamma\tau} \left[\left(1 - \frac{i\gamma}{2\omega_R} \right) e^{i\omega_R\tau} + \left(1 + \frac{i\gamma}{2\omega_R} \right) e^{-i\omega_R\tau} \right] \right\} [1 + \langle \hat{\sigma}_3(t_0) \rangle] \\ &+ \frac{E^*}{2\omega_R} e^{-(3/2)\gamma\tau} (e^{i\omega_R\tau} - e^{-i\omega_R\tau}) \langle \hat{\sigma}_-(t_0) \rangle. \end{aligned} \quad (36)$$

Carrying out Fourier transformation of Eq. (36), we will get three peaks in the resonance fluorescence spectrum.

The above comparison of the resonance fluorescence spectrum of excitons with that of a two-level atom driven by coherent light clearly illustrates that the absence of Rabi

splitting in the exciton resonance fluorescence spectrum indeed comes from the exciton's bosonic character.

To check the self-consistency of our small fluctuation approximation, we calculate the incoherent part of the total exciton number in the steady state $\bar{n} = \langle \delta\alpha^+ \delta\alpha \rangle_s$. The result

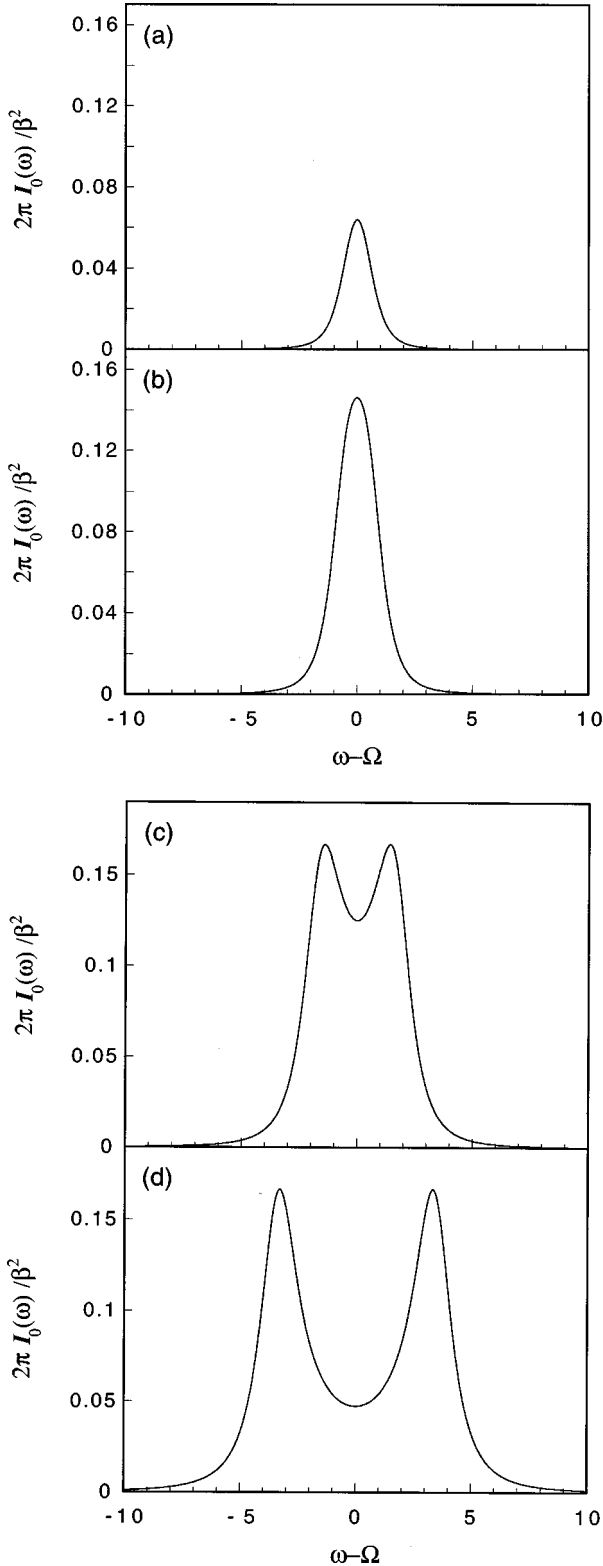


FIG. 1. The spectrum of resonance fluorescence. The value of $G^2 n^2 / \gamma^2$ is (a) 0.1, (b) 0.2, (c) 0.5, (d) 1.0. The frequency is in the unit of γ . Ω , exciton frequency; β , the parameter in Eq. (14).

may be obtained from matrix C

$$C = \begin{pmatrix} \langle \delta\alpha^2 \rangle & \langle \delta\alpha \delta\alpha^* \rangle \\ \langle \delta\alpha^* \delta\alpha \rangle & \langle \delta\alpha^{*2} \rangle \end{pmatrix} = \frac{1}{2\pi} \int_{-\infty}^{+\infty} S(\omega) d\omega. \quad (37)$$

The above integration can be carried out by contour integration easily, and the result is

$$C = \begin{pmatrix} -iG\alpha_0^2(\gamma - i4Gn) & 2G^2 n^2 \\ 2G^2 n^2 & iG\alpha_0^{*2}(\gamma + i4Gn) \end{pmatrix} \times \frac{1}{\gamma^2 + 12G^2 n^2}. \quad (38)$$

Hence,

$$\bar{n} = C_{21} = \frac{2G^2 n^2}{\gamma^2 + 12G^2 n^2}. \quad (39)$$

We see that the small fluctuation approximation holds either

$$n \gg \frac{1}{6} \quad (40)$$

or

$$\gamma^2 \gg 2G^2 n. \quad (41)$$

V. QUANTUM FLUCTUATION OF THE RADIATION INTENSITY

The directly measurable quantum fluctuation of intensity is given by

$$\sigma = \frac{\langle \hat{E}^{(-)2} \hat{E}^{(+2)} \rangle - \langle \hat{E}^{(-)} \hat{E}^{(+)} \rangle^2}{\langle \hat{E}^{(-)} \hat{E}^{(+)} \rangle^2} = \frac{\langle \hat{b}^{+2} \hat{b}^2 \rangle - \langle \hat{b}^+ \hat{b} \rangle^2}{\langle \hat{b}^+ \hat{b} \rangle^2}. \quad (42)$$

It can be evaluated by the positive P representation of the exciton density operator as

$$\sigma = \frac{\langle \alpha^{+2} \alpha^2 \rangle - \langle \alpha^+ \alpha \rangle^2}{\langle \alpha^+ \alpha \rangle^2}. \quad (43)$$

Up to the second order of $|\delta\alpha/\alpha_0|$, we get

$$\sigma = 2 \frac{\langle \delta\alpha^+ \delta\alpha \rangle}{\alpha_0^+ \alpha_0} + \frac{\langle \delta\alpha^2 \rangle}{\alpha_0^2} + \frac{\langle \delta\alpha^{+2} \rangle}{\alpha_0^{+2}} + \frac{\langle \delta\alpha^+ \delta\alpha \rangle}{(\alpha_0^+ \alpha_0)^2}. \quad (44)$$

Hence from Eq. (38), we get

$$\sigma = -\frac{2G^2(2n-1)}{\gamma^2 + 12G^2 n^2}, \quad (45)$$

which shows the fluorescence is of the nonclassical character provided $n > \frac{1}{2}$. The minimum of σ given by Eq. (45) is at

$$n = \frac{1}{2} \sqrt{1 + \frac{\gamma^2}{3G^2}}$$

We also see that when $G=0$, σ becomes 0, which is consistent with the result of only coherent elastic “scattering.”

VI. SUMMARY

In this paper, we study the effect of exciton-exciton interaction on the exciton resonance fluorescence. We demonstrate that without exciton-exciton interaction, the exciton has no resonance fluorescence due to its bosonic nature. The exciton-exciton interaction switches on resonance fluorescence, and the fluorescence spectrum is split into two peaks when the pumping field exceeds a critical value. However, there is still no Rabi splitting in the spectrum of resonance

fluorescence. The quantum fluctuation of the fluorescence light intensity is also studied. The fluorescence light exhibits nonclassical statistical property when the mean exciton number is larger than 1/2.

Our approach is based on the master equation for the density operator of the excitons, and the small fluctuation approximation is used to linearize the stochastic differential equations. A discussion on the self-consistency of this approximation is also presented.

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