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Nonlinear Optical Response of Excitons in Semiconductor Microcavities

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Excitons in semiconductor microcavities show a great variety of nonlinear optical responses depending upon the degree of fluctuation in well-thickness. A unified theory is developed to explain (1) collapse of Rabi-splitting, (2) appearance of multiplet structures against increase of pump power and (3) emission enhancement due to weak localizations of exciton polaritons.

1. INTRODUCTION

Excitons have been proved[1] theoretically and experimentally to show large third-order optical responses and rapid radiative decay, consequently resulting in large figure-of-merit far beyond the conventional limitation[2]. This is, however, limited to good crystals at low temperature where coherent length of an exciton is long enough. This limitation may be partially removed by putting these excitonic materials in a microcavity. In the present paper, we present a unified theory on a great variety of nonlinear optical responses due to excitons in semiconductor microcavities. Large Rabi-splitting and Rabi-oscillation were observed under nearly resonant conditions of cavity radiation mode and excitons in GaAs quantum wells[3, 4]. Fluctuation of well-thickness is inevitable at the present technology of semiconductor manipulation. Then the exciton level has the inhomogeneous broadening Δ. Depending upon relative value of bare Rabi-frequency g_0 to Δ, we expect different nonlinear optical responses due to excitons. In the case of such a quantum well as $g_0 >> \Delta$, the Rabi-splitting is observed to be several meV under weak pumping but it collapses into a single central mode both in transmission and emission spectra when the pump power increases above a critical value[5]. On the other hand, such a small effective Rabi-splitting as 0.3meV was observed for the similar system under large inhomogenuities $\Delta >> g_0$ (bare Rabi-frequency): In this case, the emission spectrum becomes to show quartet structure under stronger pumping while it is a doublet one for weak pumping[6]. The intermediate region is also interesting. For such a quantum well system as thickness fluctuations work as effective elastic scattering centers for excitonic polaritons, weak localization looks to induce interesting nonlinear optical responses[7]. In Section 2 to 4, we discuss linear and nonlinear optical responses in two limiting cases from unified point of view. In Section 5, the effect of weak localization on nonlinear optical responses is discussed.

2. MODEL OF SEMICONDUCTOR MICROCAVITY

Excitons in microcavity play important roles in enhancing nonlinear optical responses. This is because the exciton has large transition dipolemoment μ as long as it is coherent[8, 9] and the radiation field has large amplitude E_0 inversely proportional to $\sqrt{V}$ the square root of the microcavity volume V. As a result, the hybridized system shows large Rabi splitting proportional to μE_0 in the absorption spectrum[3] and the emission spectrum. Excitons and photons are considered both as Bose particles in the case of small number of photons and excitons. Then excitons do not show any nonlinear optical response. We discuss how to understand a great variety of nonlinear optical responses from the excitons in a microcavity. A vacuum Rabi-splitting of exciton has been observed in absorption or emission spectrum under weak pumping for a GaAs quantum well embedded in a microcavity with distributed Bragg reflectors on both ends. However, the magnitude of the splitting is ranging from 0.3 to 5meV even for a single quantum well. Furthermore, a great variety of nonlinear optical responses have been observed depending upon the sample quality of a GaAs quantum well. In this Section, a model to answer these questions are presented.

We introduce the model Hamiltonian to describe the energy fluctuation of the lowest exciton in a quantum well mainly due to the spatial fluctuation of the well thickness. The spatial extent of this fluctuation is assumed to be much larger than an exciton Bohr radius. Thus we can introduce the wavelet model described by the following Hamiltonian:

$$
\begin{aligned}
H = & \hbar\omega_c a^\dagger a + \sum_{\sigma k} \hbar\omega_k(R) b^\dagger_{\sigma k} b_{\sigma k} \\
& + \frac{1}{2}\sum_{\sigma\sigma'}\sum_{kk'} \hbar V_q^{\sigma\sigma'} b^\dagger_{\sigma k+q} b^\dagger_{\sigma' k'-q} b_{\sigma' k'} b_{\sigma k} \\
& - i\sum_{\sigma k} \hbar g_k (b^\dagger_{\sigma k} a - a^\dagger b_{\sigma k}) \\
& + i\sum_{\sigma k} \hbar h_k (b^\dagger_{\sigma k} b^\dagger_{\sigma k} b_{\sigma k} a - a^\dagger b^\dagger_{\sigma k} b_{\sigma k} b_{\sigma k}). \qquad (1)
\end{aligned}
$$

For a microcavity with a size of relevant wavelength, we may consider a single photon-mode $(a, a^\dagger)$ which is well confined by the distributed Bragg reflectors. Note that the exciton energy $\omega_k(\boldsymbol{R})$ depend on the local coordinate $\boldsymbol{R}$ of the center-of-mass motion of the exciton $(b_{\sigma k}, b^\dagger_{\sigma k})$ within the quantum well. We have two kinds of exciton nonlinearities: the third term of Eq.(1) describes the exciton-exciton collision mainly due to electron-exchange between two excitons[10], and the fifth term of Eq.(1) the state filling effect[10]. The Rabi-coupling constant g_k has dominant contribution from $k=0$ term for the system with negligible inhomogenuity. Then the coupled equations of motion for the cavity photon and the 2D exciton $b_{\alpha k}$ are derived as follows:

$$\frac{da}{dt}=-i\omega_c a+\sum_{\alpha k} g_k b_{\alpha k}-\sum_{\alpha k} hk b^{\dagger}_{\alpha k} b_{\alpha k} b_{\alpha k}-\kappa a, \quad (2)$$

$$\frac{db_{\alpha k}}{dt}=-i\omega_k(R)b_{\alpha k}-i\sum_{\sigma' k',q} V_q^{\sigma\sigma'} b^{\dagger}_{\sigma' k'} b_{\sigma' k'-q} b_{\alpha k+q}$$
$$-g_k a+h_k(2b^{\dagger}_{\alpha k} b_{\alpha k} a-a^{\dagger} b_{\alpha k} b_{\alpha k})-\gamma_k b_{\alpha k}. \quad (3)$$

Here we have introduced the decay rate 2κ of photons from the microcavity and the exciton dephasing rate γ_k. Bare Rabi frequency is given by g_0 and we assume the transition frequency $\omega_0(R)$ to obey Gaussian distribution with a width 2Δ. Depending upon the relative values of g_0, Δ, κ and γ_0, we have quite different nonlinear optical responses of this coupled system.

3. COLLAPSE OF RABI-SPLITTING

First, we discuss the case of $g_0 >> \Delta$, γ_0 and κ, which may correspond to experimental situation at University of Arizona[5]. Here assuming only exciton at $k=0$ to interact with the cavity photon, we solve the coupled equations of Green functions of $G_{aa^{\dagger}}(\omega)$ and $G_{b^{\dagger}a}(\omega)$. The common denominator of these Green functions gives eigen-energies and spectrum broadenings of the coupled system. The eigen-energies are obtained as solutions of the following equation:

$$\omega_c-\omega=\frac{(g_0-2h_0n_0)^2(\omega_0-\omega)}{(\omega_0-\omega)^2+(\gamma_0+\Gamma_{exc})^2}, \quad (4)$$

which is obtained by putting the real part of denominator to be zero. Here Γ_{exc} denotes the relaxation rate of $k=0$ exciton due to the collision with other excitons, which comes through the second term of Eq.(3) and is approximated by a constant times the total concentration of excitons. The state-filling effect $-2h_0n_0$ in Eq.(4) originates from the last term of Hamiltonian (1) or the fourth term of Eq.(3), and has been evaluated in the lowest order. The imaginary part of the denominator:

$$\Gamma(\omega)\equiv\kappa+\frac{(g_0-h_0n_0)^2(\gamma_0+\Gamma_{exc})}{(\omega-\omega_0)^2+(\gamma_0+\Gamma_{exc})^2}, \quad (5)$$

gives the broadening of each eigen-energy at $\omega=\omega_{\pm}$, i.e., the solution of Eq.(4). As Figure 1(a) shows, two eigen energies are obtained as cross-points of the left- and right-hand sides of Eq.(4) at such a low pumping as $\Gamma_{exc} << \gamma_0$. First, even when we increase the pumping power to such a degree as $\Gamma_{exc} < g_0$, the eigen-energies are fixed almost independent of pumping power as Figure 1(b) shows but both lines at $\omega_{\pm}$ are a little broadened by the second term of Eq.(5). Second the solution at origin $\omega=\omega_0=\omega_c$ has large relaxation constant $\Gamma(\omega_0)=\kappa+(g_0-h_0n_0)^2/(\gamma_0+\Gamma_{exc})$ so that it is too much broadened to be observed in the absorption and emission spectra as long as $g_0 >> (\gamma_0+\Gamma_{exc})$. Third, however, when the pumping power is increased so as to satisfy the inequality

$$\Gamma_{exc}+2\sqrt{2}h_0n_0>\sqrt{2}g_0-\gamma_0 \quad (6)$$

the Rabi-splitting disappears abruptly. Here note that both Γ_{exc} and h_0n_0 increase in linearly proportion to the exciton density in the lowest order approximation. Fourth, at the same time, the central line at $\omega=\omega_0$ becomes a little sharper and the absorption or emission structure becomes observable at such a high pumping as Eq.(6) is satisfied. These four features from the first to fourth items are well coincident to the observations at Arizona. Fifth, when the detuning of $|\omega_0-\omega_c|$ is increased under a fixed pumping rate, the Rabi-splitting is also expected to collapse as Figures 1(b) and (c) show.

4. APPEARANCE OF BIEXCITON STRUCTURE

We have quite different situation when the exciton transition frequency $\omega_0(\boldsymbol{R})$ depends on the spatial coordinate $\boldsymbol{R}$. Here we imagine the large fluctuation of the quantum-well thickness and the exciton system to be confined to one of the two-dimensional islands with the different energies. Then the exciton coherency is limited to the size of islands as in the microcrystallite[8]. The Gaussian distribution is assumed for these exciton-energies with the characteristic width 2Δ. Let us study first the linear response of Rabi-splitting and then the nonlinear one for the second case when $\Delta>\kappa, g_0, \gamma_0$ which corresponds to experimental situation at Stanford[6]. As shown in Figure 2, we can point out that the effective Rabi splitting becomes less than the bare Rabi splitting $2g_0$. Although whole the excitonic transitions look to contribute to the Rabi-splitting in the operator form from the second term of Eq.(2), only a small part of excitonic transitions near $\omega_0=\omega_c$ can contribute effectively to the splitting. Thus the effective Rabi-splitting should be determined self-consistently depending upon the relative value Δ/g_0.

We will try to understand this reduction of the effective Rabi-splitting from a different view point. The quantum well consists of many islands accommodating the exciton with the different energies and the exciton is almost confined within one of these islands. The nonlinear optical responses of this coupled system reflect such a situation, and quite different from the first case of Section 3. The average size of these islands is denoted by S_{coh} and then the quantum well may be considered to consist of approximately S/S_{coh} two-dimensional microstructures, where S is the effective area of the quantum well. We may imagine S_{coh} to be of an order of magnitude ranging from several times to several hundred times πa_b^2 with a_b

the exciton Bohr radius. Then under strong pumping, one of these islands become to contain a few excitons. Here we assume that these excitons interact with each other within the island but not directly with those in other islands while they interact with each other only through the radiation field within the microcavity. We have solved the eigen-energies and eigen-vectors of one-exciton and two-excitons states within a single island, which interacts strongly with the radiation field in the microcavity. Here we assumed that two excitons with opposite spin structures interact attractively and make a bound state of biexciton but neglected the repulsive one among the same-spin excitons[9]. In terms of these solutions, we calculate the emission spectrum and the time-dependent Rabi-oscillation[3]. Then we can understand the doublet and quartet structures in the emission spectrum, respectively, at low and intermediate pumpings. Fourier transform of these signals can also explain the Rabi-oscillation and the stronger oscillation in time, also respectively at low and intermediate pumpings.

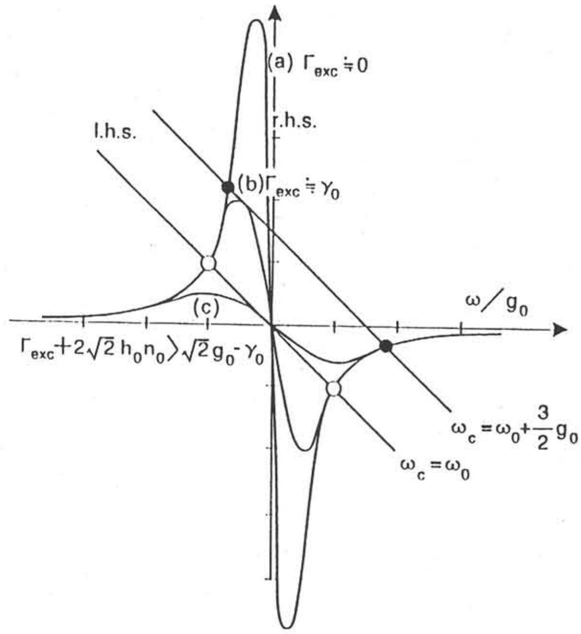

Figure 1. Rabi-splitting of homogeneous exciton. The left- and right-hand sides of Eq.(4) are plotted against ω/g_0. (a) For such a week pumping as $\Gamma_{exc} << \gamma_0$, (b) intermediate pumping $\Gamma_{exc} \sim \gamma_0$, and (c) so strong pumping $\Gamma_{exc} + 2\sqrt{2}h_0 n_0 > \sqrt{2}g_0 - \gamma_0$ that the Rabi-splitting is collapsed. The energy separation between two white circles and that of black ones denote the Rabi-splitting, respectively, under the resonant $\omega_c = \omega_0$ and the off-resonant case $\omega_c = \omega_0 + g_0$.

5. WEAK LOCALIZATION

Fluctuations in quantum-well thickness work as scatterers of excitonic polaritons (EPs) in the intermediate regions. When we pump the EPs at $\boldsymbol{k}_0$, they are most effectively backward scattered into $-\boldsymbol{k}_0$ by weak localization due to those thickness fluctuations. In the semiconductor microcavity, the state-density of EPs becomes maximum at two-dimensional wavevector $\boldsymbol{k} = 0$ which are propagating perpendicular to the quantum-well. As a result, this system shows laser action most easily in this direction and two EPs with $\boldsymbol{k}_0$ and $-\boldsymbol{k}_0$ are scattered into the laser mode by induced scatterings[7]. For detailed theory of these processes, see ref.11).

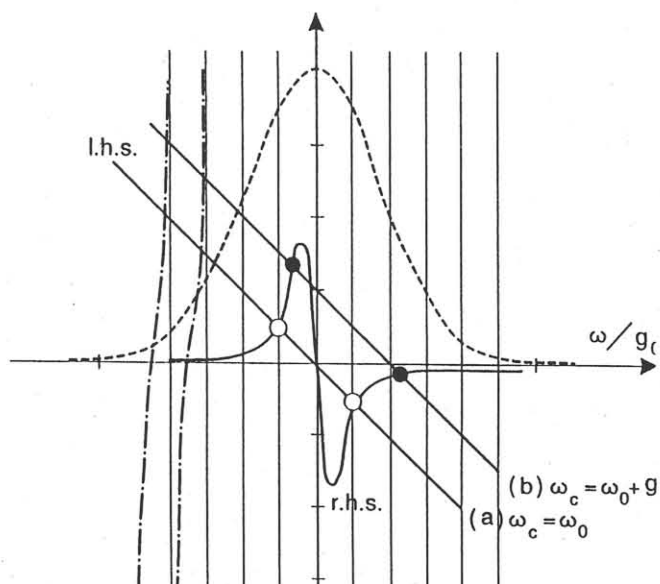

Figure 2. Rabi-splitting of inhomogeneous exciton. Sketch of left- and right-hand sides of Eq.(7). Dotted curve the Gaussian distribution of exciton level.

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