Parametric Scattering of Cavity Polaritons

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(Received 16 April 1997)

We report frequency-domain degenerate four-wave mixing results on excitons in a semiconductor microcavity in the strong coupling regime. The excitation is kept to the $\chi^{(3)}$ limit. The spectral response shows strong polarization dependence. We analyze the data with a new model where the degenerate four-wave mixing process is described as an elastic scattering of two cavity polaritons mediated by the state filling effect and the two-body attractive and repulsive interaction between excitons. From the polarization and detuning dependence, the ratio of the coupling constants of these three terms is uniquely determined. We find that the attractive interaction term and the state filling are the dominant contributions to the nonlinearity. [S0031-9007(97)03813-1]

PACS numbers: 71.36.+c, 42.50.–p, 42.65.–k, 71.35.Lk

The exciton-photon coupled system in a semiconductor microcavity structure is attracting a lot of current interest in both fundamental and applied research [1]. In the strong coupling regime very large normal mode splitting exists which is several orders of magnitude larger than that of the atomic system [2]. Various experimental and theoretical studies have been performed to elucidate the features characteristic to the excitonic system [2–7]. As long as linear optical responses are concerned the semiclassical picture with anomalous dispersion associated with excitons can well explain these features as it was pointed out for the atomic system [8]. In the nonlinear regime, the atom-cavity coupled system shows characteristic features originating from the granular nature of the radiation field [9]. Because of the similarities with the atomic system and ease of integration with the existing semiconductor technology, the exciton-cavity coupled system is expected to be suitable for the fabrication of quantum logic devices. However, the atomic two-level system can be saturated with as little as one photon and shows a large nonlinearity, whereas the excitonic system behaves mostly harmonic in the low density regime. Any deviation from this harmonic behavior is due to the nonideal bosonic nature of excitons. This deviation is essential to photon manipulation. Therefore, elucidation of the nonlinear optical responses of the exciton-cavity coupled system is crucial to the realization of the idea of quantum logic devices.

In the conventional scheme of nonlinear optics the nonlinear polarization is evaluated as a higher-order perturbation of the exciton-photon dipole interaction. This framework is not applicable to the strong coupling regime, because the exciton-photon dipole interaction is fully taken into account nonperturbatively in order to form cavity polaritons. Nonlinear responses are described as higher-order scattering of cavity polaritons by the anharmonicities which originate from the fermionic nature and Coulomb interaction of the constituents of the excitons. Nonlinear optical processes of bulk polaritons should also be treated in the same framework. For the bulk case, however, the propagation effects of polaritons bring further complications. Therefore, studies on the nonlinear response of the cavity-exciton coupled system can help us with a deeper understanding of the polariton concept itself and bring up a new aspect of nonlinear optics.

We will show in this Letter that the widely accepted third-order perturbation theory fails to describe the nonlinearities of the exciton-photon coupled system even when the excitation is strictly kept to the lowest-order power level of the $\chi^{(3)}$ limit. A new model that is able to account for the experimental observations will be presented.

The semiconductor microcavity sample used in this work consists of a 12-nm-thick single quantum well (SQW) placed at the antinode of a $\lambda/2$-planer microcavity with an AlAs spacer sandwiched between the two GaAs/AlAs Bragg reflectors (22 and 14.5 pairs for bottom and top, respectively). We scan the detuning energy $\Delta \equiv \hbar\omega_c - \hbar\omega_e$ by selecting the spot position on the sample which has a tapered cavity length across the surface.

We perform degenerate four-wave mixing (DFWM) experiments in the self-pumped phase conjugation geometry [10] using tunable picosecond pulses (pulse width of 1.9 psec and spectral width of 0.7 meV) from a mode locked Ti:sapphire laser. We can tune the center frequency of the pulses continuously without losing mode locking. The recorded DFWM signals, as a function of laser frequency, are called frequency-domain DFWM (FD-DFWM) spectra. The pump beam is directed perpendicular to the sample surface, while the test beam is slightly tilted (about 3°) from normal incidence. The pulse energy of the beams is kept low so that the exciton density is less than 10^9 cm^2. At this excitation level, FD-DFWM signals are linear to the test beam intensity and square to the pump beam intensity. No power dependent change of the spectral response is observed.

The dotted lines of the bottom graph in Fig. 1 show the linear reflection spectra at various spot positions on the sample, and therefore various detunings. Two dips associated with the heavy-hole exciton resonance and cavity resonance are observed. The upper graph shows the spot of normal mode resonance as a function of spot position. The calibrated detuning energy is indicated at the abscissa. The mode splitting at zero detuning is 4.3 meV which is larger than the linewidths of the exciton or the cavity at about 1.5 meV. Solid lines in the bottom graph show FD-DFWM spectra in the co-circularly polarized configuration, where polarization states of the pump and test beam are σ_+. Strong signals appear at both the upper and lower branch resonance. We perform the FD-DFWM experiments in the following polarization configurations: A (x, x, x), B (x, y, y), C $(\sigma_+, \sigma_+, \sigma_+)$, D (x, σ_+, σ_+), E (x, σ_+, σ_-), where the polarization states in a bracket denote pump beam, test beam, and signal beam, respectively. Figure 2 shows FD-DFWM spectra at $\Delta = 0$ for all polarization configurations. A strong signal appears at the upper branch resonance in the parallel configuration (A) while the lower branch signal dominates in the orthogonal configuration (B). In the other polarization configurations (C, D, and E), signals appear in both the upper and lower modes.

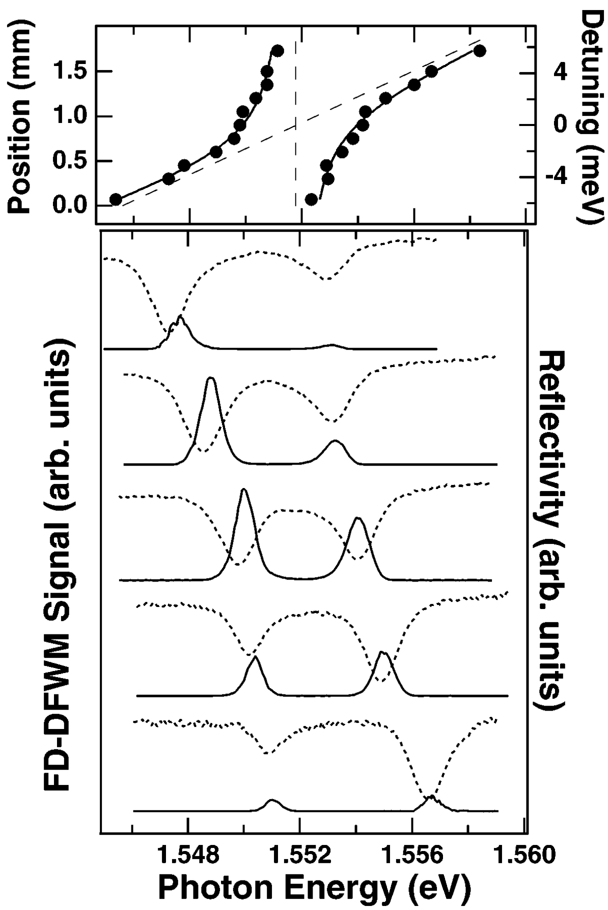

FIG. 1. Linear reflection spectra (dotted lines) and degenerate four-wave mixing spectra (solid lines) of the GaAs SQW for various detunings. DFWM measurements are performed in the co-circular polarization configuration, where all the beams are σ_+ polarized. In the upper graph, the normal mode resonance observed in linear reflection spectra is plotted as a function of spot position. The calibrated energy scale for detuning energy is shown.

In our previous paper [10], we analyzed the effective field amplitude at the position of the SQW in the microcavity and evaluated the enhancement factor for the phase conjugation reflectivity by considering the exciton-photon dipole interaction a perturbation. In the strong coupling limit the internal field amplitude disappears at the bare exciton resonance, and strong suppression of the DFWM signal is expected. This contradicts our experimental results. Therefore, in order to explain the observed features, we need a new picture for DFWM. We show in the following that a model based on cavity-polariton concept can account for the experimental results.

First, we consider the unperturbed Hamiltonian H_0. The helicity, which is the z component of the angular momentum J_z, is a good quantum number, and thus we take circularly polarized states, $J_z = 1$ (σ_+) or $J_z = -1$ (σ_-) as the bases. In these bases H_0 is expressed as

$$H_0 = \sum_{\sigma=\pm} [\hbar\omega_c a_\sigma^\dagger a_\sigma + \hbar\omega_e b_\sigma^\dagger b_\sigma + g(a_\sigma^\dagger b_\sigma + b_\sigma^\dagger a_\sigma)], \qquad (1)$$

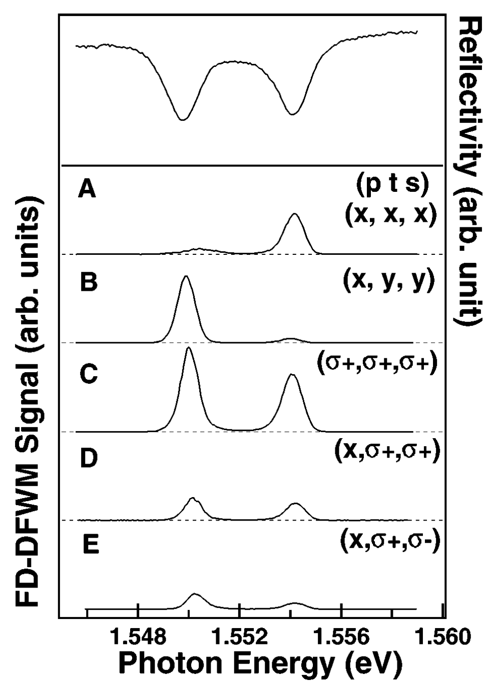

FIG. 2. Spectra of FD-DFWM at zero detuning for various polarization configurations indicated in the figure are described in the text.

where a_σ and b_σ denote the annihilation operators of the cavity photons and excitons with helicity σ and frequencies ω_c and ω_e, respectively. We assume that the cavity photons and excitons have no lateral momentum, i.e., $\vec{k}_\parallel = 0$, and we omit writing the wave vector indices. The constant g expresses the strength of the exciton-photon coupling. We can diagonalize H_0 with cavity-polariton modes,

$$H_0 = \sum_{\sigma=\pm} [\hbar\omega_\alpha \alpha_\sigma^\dagger \alpha_\sigma + \hbar\omega_\beta \beta_\sigma^\dagger \beta_\sigma], \quad (2)$$

where α_σ (β_σ) is the annihilation operator of the upper (lower) branch mode with resonance frequencies ω_α (ω_β). The linear coupling coefficients of cavity photon components and exciton components are a function of the detuning Δ. Please note that photon and exciton components are out of phase for the upper polariton mode and in phase for the lower polariton.

Figure 3 shows a schematic of the self-pumped phase conjugation geometry. The pump beam excites two counterpropagating polaritons. The higher-order coupling between the excitons and the cavity photons, which is expressed with the nonlinear Hamiltonian H_{nl}, causes the elastic scattering shown in Fig. 3(b). The test beam selects the modes of the outgoing polaritons. The backward-going polariton is connected to the phase conjugation signal photon outside of the cavity. When the nonlinear interaction is much weaker than the exciton-photon coupling, we can treat H_{nl} as a perturbation within the interacting boson picture. The FD-DFWM signal intensity at the polariton resonance is proportional to the scattering probabilities I_{PC}, which can be expressed as [11,12]

$$I_{\mathrm{PC}} \propto \begin{cases} |\langle \alpha_{4\sigma_4}\alpha_{3\sigma_3}|H_{\mathrm{nl}}|\alpha_{2\sigma_2}\alpha_{1\sigma_1}\rangle|^2 |\langle \alpha \mid a\rangle|^8 I_1 I_2 I_3 \\ \quad (\text{for } \omega = \omega_\alpha) \\ |\langle \beta_{4\sigma_4}\beta_{3\sigma_3}|H_{\mathrm{nl}}|\beta_{2\sigma_2}\beta_{1\sigma_1}\rangle|^2 |\langle \beta \mid a\rangle|^8 I_1 I_2 I_3 \\ \quad (\text{for } \omega = \omega_\beta)\,. \end{cases} \quad (3)$$

Here, the I_i's are the intensity of the ith beam. The beam indices, $i = 1, 2, 3$, and 4, denote the beams of forward pump, backward pump, test, and signal, respectively. In the scattering processes the total J_z of the two incoming and outgoing polaritons are conserved. H_{nl} has the following terms:

$$H_{\mathrm{nl}} = H_1 + H_2 + H_3 + \mathrm{H.c.}, \quad (4)$$

$$H_1 = W(b_{4+}^\dagger b_{3-}^\dagger b_{2+} b_{1-} + b_{4+}^\dagger b_{3-}^\dagger b_{2-} b_{1+} + b_{4-}^\dagger b_{3+}^\dagger b_{2+} b_{1-} + b_{4-}^\dagger b_{3+}^\dagger b_{2-} b_{1+}), \quad (5)$$

$$H_2 = R \sum_{\sigma=\pm} b_{4\sigma}^\dagger b_{3\sigma}^\dagger b_{2\sigma} b_{1\sigma}\,, \quad (6)$$

$$H_3 = -g\nu \sum_{\sigma=\pm} [b_{4\sigma}^\dagger b_{3\sigma}^\dagger b_{2\sigma} a_{1\sigma} + b_{4\sigma}^\dagger a_{3\sigma}^\dagger b_{2\sigma} b_{1\sigma}]. \quad (7)$$

The first term represents the interaction between two excitons with opposite helicity. Resonant two polariton scattering via a biexciton state with $J_z = 0$ is related to this term [12]. The second and third term act on pairs of polaritons with the same helicities. The term H_2 represents the repulsive interaction between two excitons with same helicity. The last term, H_3, represents the reduction of the exciton-photon coupling due to phase space filling [13,14]. The occupation of the exciton with density $\langle b^\dagger b\rangle$ changes the exciton-photon coupling g to $g(1 - \nu\langle b^\dagger b\rangle)$ ($\nu > 0$). In the configurations C and D the test beam and signal beam have the same helicity, and thus H_2 and H_3 are effective. In the E configuration the two outgoing polaritons have opposite helicities, and only H_1 contributes to the nonlinear scattering. In the cases of the linear polarization configurations A and B, the one-exciton state is expressed as $|b_{\mathbf{k}X}\rangle = (|b_{\mathbf{k}+}\rangle + |b_{\mathbf{k}-}\rangle)/\sqrt{2}$ or $|b_{\mathbf{k}Y}\rangle = (|b_{\mathbf{k}+}\rangle - |b_{\mathbf{k}-}\rangle)/\sqrt{2}\,i$. The intermediate two polariton states are the linear combination of the $J_z = 0$ state and $J_z = \pm 2$ state, and all terms contribute to the DFWM signal. In order to understand the mechanism of the mode switching, we examine the role of the relative phase of the exciton and photon component in the upper (α) and the lower (β) mode operators. The terms H_1 and H_2 consist of only exciton operators with the scattering amplitudes for both modes having the same sign. On the other hand, the phase space filling terms in H_3 have one photon operator and three exciton operators, and therefore the scattering amplitudes show opposite sign for the upper and lower modes. In the parallel configuration (A), the signals at the upper and lower mode resonance at zero detuning are

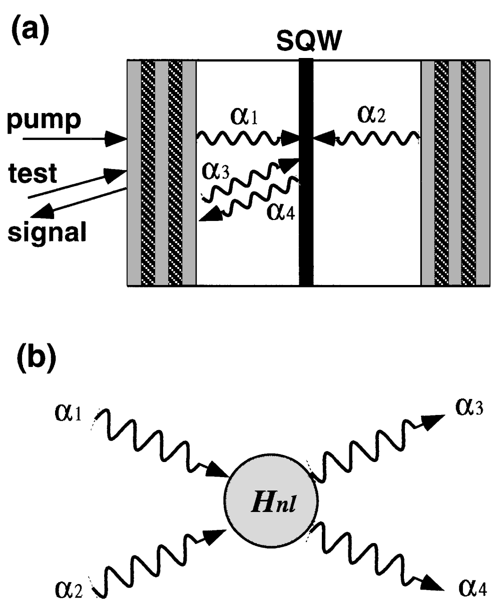

FIG. 3. (a) Schematic of the self-pumped phase conjugation experiment. (b) Relevant processes for DFWM based on the cavity-polariton picture. Two incoming polaritons provided by the pump beam are elastically scattered into two outgoing polaritons conserving energy and momentum.

expressed as $I_{PC}(\omega = \omega_\alpha; A) \propto |W/4 + (R/2 - g\nu)|^2$ and $I_{PC}(\omega = \omega_\beta; A) \propto |W/4 + (R/2 + g\nu)|^2$. When we change the test beam polarization from an x to a y direction, the relative signs of the $J_z = 0$ term and $J_z = \pm 2$ terms change. The signals in the orthogonal configuration (B) are $I_{PC}(\omega = \omega_\alpha; B) \propto |W/4 - (R/2 - g\nu)|^2$ and $I_{PC}(\omega = \omega_\beta; B) \propto |W/4 - (R/2 + g\nu)|^2$. The fact that the upper branch signal of (A) is switched to the lower mode in (B) invokes the conditions, $|W/4|, |-g\nu| > |R/2|$, and $W < 0$. The negative sign of W indicates that the interaction between σ_+ and σ_- excitons is attractive irrespective of the existence of a stable biexciton state.

Figure 4(a) shows the experimental data for the detuning dependence of the upper and lower mode signals for all polarization configurations. We calculate the corresponding signal intensities for various values of R, W, and $g\nu$. We find that the ratio of these three parameters is uniquely determined in order to match the theoretical curves to the experimental data. Figure 4(b) shows the calculated curves for the ratio as $W/4 : R/2 : g\nu = -0.75 : 0.1 : 1.0$. The spectra of Fig. 2 are well reproduced with the same parameters [11].

In conclusion, we have reported mode and polarization dependent degenerate four-wave mixing in a semiconductor microcavity structure in the strong coupling regime. The observed features clearly show the failure of the standard framework of third-order perturbation of the exciton-photon dipole interaction. We propose a new treatment, where FD-DFWM is described as stimulated parametric scattering of cavity polaritons. The obtained agreement between theory and experiment is excellent. Scattering is caused by anharmonicities in the interaction potential resulting from the attractive and repulsive interaction between excitons and phase space filling. The attractive interaction and phase space filling effect dominantly contribute to the excitonic nonlinearity in GaAs quantum well systems.

Systematic experiments which involve changing the exciton-photon coupling are necessary in order to find the way to the present cavity-polariton model from conventional nonlinear optics.

The authors would like to thank Dr. Georg Mohs for the critical reading of the manuscript. This work was supported by a grant-in-aid for scientific research in the priority area of Mutual Quantum Manipulation of Radiation Field and Matter from the ministry of Education, Science and Culture of Japan and the CREST (Core Research for Evolutional Science and Technology) by the Japan Science and Technology Corporation (JST).

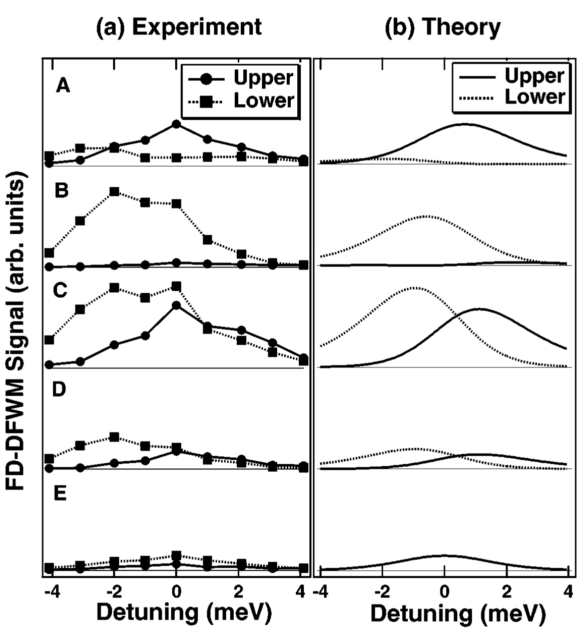

FIG. 4. Detuning dependence of the DFWM signal for the upper and lower mode resonance for the same polarization configurations as in Fig. 2. (a) Experimental data and (b) calculated curves.

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