Light squeezing via a biexciton in a semiconductor microcavity

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(Received 24 August 2010; revised manuscript received 9 November 2010; published 21 January 2011)

We theoretically investigate the squeezing of light by the interaction between a biexciton and cavity polaritons. As a model system, we consider a CuCl-quantum-well microcavity in which a biexciton can stably exist. An effective biexciton-polariton coupled state can be formed by controlling the polariton level structure by modifying the quantum-well thickness. We analyze in detail the nonlinear optical response obtained from the coupled state, and evaluate the light squeezing by directly calculating the variance of quantum noise in the output field, in terms of the dependence of the squeezing on the incident light intensity. We clarify an optimal condition yielding maximum squeezing and show that squeezing is enhanced by utilizing biexciton nonlinearity, compared with the squeezing achieved in a system having no biexciton.

DOI: 10.1103/PhysRevB.83.045305

PACS number(s): 71.36.+c, 42.50.Dv, 42.50.Pq

I. INTRODUCTION

Light squeezing can be realized in various nonlinear optical processes such as parametric down-conversion, parametric amplification, and degenerate four-wave mixing (FWM). For device applications, the generation of squeezed light in semiconductors is desirable though it becomes more difficult to achieve light squeezing in semiconductors because of thermal noise. However, recent development of semiconductor microcavities enables us to create a cavity polariton, in which an exciton is strongly coupled to a cavity photon so that the exciton-photon coupling can overcome thermal noise. Cavity polaritons allow us to achieve efficient light squeezing because of the strong optical nonlinearity of excitons. In fact, polariton parametric amplification¹ and polariton degenerate FWM² have been experimentally demonstrated, and polariton squeezing by using semiconductor microcavity systems has recently been extensively investigated.3-5

Most of the above polariton-squeezing methods are based on polariton-polariton scattering. To our knowledge, there are only a few methods that exploit the optical nonlinearity of a biexciton. This is because a biexciton is generally unstable, and the implementation of an exciton-cavity system with a stable biexciton is quite difficult. Recently, however, a CuCl-quantum-well microcavity system has been reported.⁶ This system has a stable biexciton and allows us to achieve an interaction between the biexciton and cavity photons in a low-dimensional nanoscale structure. Further, it is possible to create a biexciton-polariton coupled state by selecting the appropriate cavity parameters and thickness and number of the quantum well. Biexciton nonlinearity can be strongly enhanced by utilizing the cavity quantum electrodynamics effect.⁷

In this study, we theoretically investigate the polariton squeezing utilizing biexciton nonlinearity. Generally, degenerate FWM is simple and suitable² in the generation of squeezed light in an exciton-cavity system. Therefore we consider a normally incident FWM as depicted in Fig. 1. Further, we focus on a CuCl-quantum-well microcavity system. In order to clarify the effect of the biexciton on light squeezing, we compare squeezing in a system having a stable biexciton with that in a system having no biexciton. We analyze in detail the

nonlinear optical response of a biexciton-polariton coupled state, and evaluate the light squeezing by directly calculating the variance of quantum noise in an output field, in terms of the dependence of the squeezing on the incident light intensity. We show that there is an optimal condition under which light squeezing is maximum, and the squeezing can be enhanced by using biexciton nonlinearity, compared with the squeezing achieved in a system having no biexciton.

The rest of this paper is organized as follows. In Sec. II, we describe an exciton-cavity system forming cavity polaritons and a biexciton, the optical master equation combined with the input-output relation, and light squeezing in the output field. In Sec. III, we analyze in detail the nonlinear optical response of the cavity polaritons and the dependence of squeezing on the incident light intensity. In Sec. IV, we summarize our results.

II. MODEL

A. One-dimensional model

As a model system, we consider an exciton system confined in a one-sided microcavity, as depicted in Fig. 1, where a $\lambda/2$ cavity is assumed and the exciton system is placed at the center of the cavity. Pump and probe beams are normally incident on the surface of the cavity and penetrate into the cavity. The incident photons are coupled to excitons, forming cavity polaritons, and are then emitted into the output field. In this study, we focus on a signal normally emitted on the same side as the incident light beams. For simplicity, in these optical processes, we ignore the nonradiative decay of excitons.

For $\hbar = c = 1$, the Hamiltonian for the fundamental mode of a $\lambda/2$ cavity field is given by

$$\hat{H}_C = \omega_c \hat{c}^\dagger \hat{c},\tag{1}$$

where $\hat{c}(\hat{c}^{\dagger})$ is the annihilation (creation) operator of the cavity photon with the energy ω_c . The interaction Hamiltonian describing the exciton-cavity coupling is given by

$$\hat{H}_{\rm int} = \sum_{k} g_k (\hat{c}^{\dagger} \hat{b}_k + \hat{c} \hat{b}_k^{\dagger}), \qquad (2)$$

where k is the quantum number of the exciton states, g_k is the exciton-cavity coupling energy, and $\hat{b}_k(\hat{b}_k^{\dagger})$ is the exciton



FIG. 1. Schematic of pump-probe assignment. k is the wave number of exciton states. g is the coupling energy.

annihilation (creation) operator. When g is larger than the spontaneous emission of excitons and the cavity damping, an exciton is strongly coupled to a cavity photon, and a cavity polariton is formed. If the spontaneous emission of excitons is negligibly small, the system can be reduced to a one-dimensional input-output system.

B. Biexciton and cavity polaritons

For an exciton system, we adopt a one-dimensional discrete-lattice model with only one degree of freedom perpendicular to the cavity surface. This model well approximates the center-of-mass motion of excitons in a CuCl and has the advantage that it provides solutions of bound and unbound two-exciton states as a complete set. Further, in this model, we ignore the spin degree of freedom of excitons, for simplicity. The Hamiltonian of the exciton system can be described as

$$\hat{H}_{X} = \epsilon \sum_{\ell} \hat{b}_{\ell}^{\dagger} \hat{b}_{\ell} - t \sum_{\ell} (\hat{b}_{\ell+1}^{\dagger} \hat{b}_{\ell} + \hat{b}_{\ell}^{\dagger} \hat{b}_{\ell+1}) + V \sum_{\ell} \hat{b}_{\ell}^{\dagger} \hat{b}_{\ell}^{\dagger} \hat{b}_{\ell} \hat{b}_{\ell} - \Delta \sum_{\ell} \hat{b}_{\ell}^{\dagger} b_{\ell+1}^{\dagger} \hat{b}_{\ell+1} \hat{b}_{\ell}, \qquad (3)$$

where $b_{\ell}(b_{\ell}^{\dagger})$ is the exciton annihilation (creation) operator at the ℓ th site, ϵ is the excitation energy of each site, and t is the transfer energy of an exciton from a site to neighboring sites. The third term is the repulsive interaction between two excitons occupying the same site. The last term is the attractive interaction between two neighboring excitons, which leads to the formation of a biexciton. By setting the boundary condition that the exciton states are zero at sites $\ell = 0$ and $\ell = N + 1$, the eigenenergy and eigenstate for the one-exciton state can be respectively given by

$$\omega_k = \epsilon - 2t \cos ka, \tag{4}$$

$$|k\rangle = \sqrt{\frac{2}{N+1}} \sum_{\ell} \sin(k\ell a) \hat{b}_{\ell}^{\dagger} |0\rangle, \qquad (5)$$

where $k = n\pi/(N+1)a$ is the discrete wave number of excitons, characterized by integer *n*, *a* is the lattice constant, and *N* is the size of the exciton system. For the repulsive exciton-exciton interaction, we consider the limit of $V \to \infty$. This prohibition of two excitons at the same site corresponds to the Pauli exclusion principle. The eigenstates for a two-exciton state can then be simply written as $|\mu\rangle = \sum_{\ell < m} C_{\ell,m}^{(\mu)} |\ell,m\rangle$, where $|\ell,m\rangle \equiv |\ell\rangle \otimes |m\rangle$ and the states of $|m,m\rangle$ for any *m* are excluded. The coefficients $C_{\ell,m}^{(\mu)}$ are numerically calculated and determined so $\{|\mu\rangle\}_{\mu}$ forms a complete system. The *k*



FIG. 2. Energy levels as a function of the size of exciton system N. (a) Energy ω_{μ} of two-exciton states $|\mu\rangle$ measured from $2\omega_{k_1}$. (b) Energy ω_{1p} of cavity-polariton states $|1p\rangle$ measured from ω_c . (c) Energy ω_{2p} of two-cavity-polariton states $|2p\rangle$ measured from $\omega_{LPB} - \omega_c$. (d) Energy ω_{2p} of two-cavity-polariton states with no biexciton measured from $\omega_{LPB} - \omega_c$. The exciton parameters are $t = 0.057 \text{ eV}, \Delta = 0.189 \text{ eV}, \text{ and } \epsilon = 3.2 + 2t \text{ eV}, \text{ leading to } \Delta_B = 0.03 \text{ eV}.$ In (b), (c), and (d), $\omega_c = \omega_{k_1}$ and g is set to $g_{k_1} = 0.004 \text{ eV}$ at N = 2. ω_{LPB} denotes the energy of LPB. Dashed line in (c) corresponds to the LPB in (b).

representation can be obtained simply by Fourier transforming $C_{\ell,m}^{(\mu)}$ to $C_{k,k'}^{(\mu)}$. Figure 2(a) shows the eigenenergies ω_{μ} of $|\mu\rangle$ as a function of *N*. The biexciton binding energy is uniquely determined by *t* and Δ . We define the binding energy of the biexciton with the lowest energy for $N \gg 1$ as Δ_B .

The eigenstates of the exciton-cavity system can now be obtained by diagonalizing the Hamiltonian, $\hat{H}_{sys} = \hat{H}_X + \hat{H}_C + \hat{H}_{int}$. Rewriting the exciton operator \hat{b}_k using the Hubbard operators as⁸

$$\hat{b}_{k} = |G\rangle\langle k| + \sum_{k'} \langle k' | \hat{b}_{k} | k, k' \rangle | k' \rangle \langle k, k' | + \cdots, \qquad (6)$$

the eigenstates can be expressed, in form, as

$$|1p\pm\rangle = \alpha_{\rm x}|k;0\rangle \pm \alpha_{\rm c}|G;1\rangle,\tag{7}$$

$$|2\mathbf{p}\rangle = \alpha_{\mathrm{xx}}|k,k';0\rangle + \alpha_{\mathrm{xc}}|k;1\rangle + \alpha_{\mathrm{cc}}|G;2\rangle, \tag{8}$$

where $|G\rangle$ is the ground state and the separation by a semicolon denotes $|exciton; photon\rangle$. $|1p-\rangle$ is the lower polariton branch (LPB) and $|1p+\rangle$ is the upper polariton branch (UPB). Figure 2(b) shows the eigenenergies ω_{1p} of $|1p\rangle$ as a function of N. $|2p\rangle$ is the two-cavity-polariton state consisting of the two-exciton state $|k,k';0\rangle$, the one-exciton-one-photon state $|k;1\rangle$, and the two-photon state $|G;2\rangle$. In this study, since we focus on the nonlinear optical response of a biexciton and cavity polaritons, cavity-polariton states up to $|2p\rangle$ are required.⁹ Figure 2(c) shows the eigenenergies ω_{2p} of $|2p\rangle$ as a function of N. For comparison, the eigenenergies for a system with $\Delta = 0$ (no biexciton) is shown in Fig. 2(d). One can find the polariton energy structure in Fig. 2(c) is drastically modified by biexciton states. The value of g_k is dependent on N through the dipole transition matrix of an exciton, $\sum_{\ell} \langle G | \hat{b}_{\ell} | k \rangle$, and it is proportional to $\sum_{\ell} \langle G | \hat{b}_{\ell} | k \rangle$. The value of $g_{\mu k}$ for $|2p\rangle$ can be calculated from $g_{\mu k} = \langle k; 1 | \hat{H}_{int} | \mu; 0 \rangle$. Thus the coupling energy is varied in accordance with N, and the polariton energy structure can also be modified by varying N. The difference between the exciton-cavity couplings in one- and two-exciton states, g_k and $g_{\mu k}$, strongly affects the strength of nonlinearity.

C. Master equation and input-output relation

The nonlinear optical response of the exciton-cavity system can be analyzed by using the optical master equation coupled with the input-output theory.¹⁰ The master equation is given by

$$\frac{d\hat{\rho}}{dt} = -i[\hat{H}_{\rm sys} + \hat{H}_{\rm ext}, \hat{\rho}] + \kappa (2\hat{c}\hat{\rho}\hat{c}^{\dagger} - \hat{c}^{\dagger}\hat{c}\hat{\rho} - \hat{\rho}\hat{c}^{\dagger}\hat{c}), \quad (9)$$

where κ is the cavity damping energy. For a CuCl microcavity system considered in this study, the spontaneous emission of excitons into noncavity modes is negligibly smaller than g, κ , and Δ_B . Therefore we ignore it in Eq. (9), for simplicity. \hat{H}_{ext} is the interaction Hamiltonian between intracavity photons and classical cw incident light beams, $\xi_{in}(t)$, given by

$$\hat{H}_{\text{ext}} = i\sqrt{2\kappa}\hat{c}\,\xi_{\text{in}}(t) + \text{H.c.},\tag{10}$$

where $\xi_{in}(t) = \xi_{pump}(t) + \xi_{probe}(t)$. For the calculation of thirdorder nonlinear optical response, we consider \hat{H}_{ext} as a perturbation term and calculate the density operator $\hat{\rho}^{(3)}$. As is well known, the optical nonlinearity is evaluated using the susceptibility χ . In exciton-cavity systems, however, there is no counterpart of χ . Therefore we directly evaluate the thirdorder output field by using the input-output theory. According to the input-output theory, the output field operator is given by

$$\hat{\xi}_{\text{out}} = \hat{\xi}_{\text{in}} + \sqrt{2\kappa}\hat{c}\hat{\rho}.$$
(11)

Using Eq. (11) with $\langle \hat{\xi}_{in} \rangle = \xi_{in}$, the third-order output field after eliminating ξ_{in} can be described as

$$\xi_{\text{out}}^{(3)} = \sqrt{2\kappa} \langle \hat{c} \rangle^{(3)}, \qquad (12)$$

where $\langle \hat{c} \rangle^{(3)} = \text{Tr}[\hat{c}\hat{\rho}^{(3)}]$. From the third-order nonlinear response, we first identify an optimal system size *N* where a strong optical nonlinearity is achieved.

D. Variance of quantum noise and light squeezing

The main interest and applications of squeezed light lie in its quadrature-operator properties. Therefore we focus on the quadrature squeezing of output field. The output field operators can be rewritten by using the quadrature operators \hat{X}_{out} and \hat{Y}_{out} as¹¹

$$\hat{\xi}_{\text{out}} = \frac{1}{2}(\hat{X}_{\text{out}} + i\hat{Y}_{\text{out}})e^{i(\theta - \omega t)}, \qquad (13)$$

$$\hat{\xi}_{\text{out}}^{\dagger} = \frac{1}{2} (\hat{X}_{\text{out}} - i\hat{Y}_{\text{out}}) e^{-i(\theta - \omega t)}, \qquad (14)$$

where θ is the reference phase and ω is the reference energy. In this study, we evaluate the squeezing by directly calculating the quadrature variances given by

$$\Delta X^2 = \langle \hat{X}_{\text{out}}^2 \rangle - \langle \hat{X}_{\text{out}} \rangle^2, \qquad (15)$$

$$\Delta Y^2 = \langle \hat{Y}_{\text{out}}^2 \rangle - \langle \hat{Y}_{\text{out}} \rangle^2.$$
(16)

Using Eqs. (11), (13), and (14), Eqs. (15) and (16) can be rewritten as

$$\Delta X^{2} = 2\kappa (2\langle \hat{c}^{\dagger} \hat{c} \rangle - 2\langle \hat{c}^{\dagger} \rangle \langle \hat{c} \rangle + 1) + \operatorname{Re}[4\kappa (\langle \hat{c} \hat{c} \rangle - \langle \hat{c} \rangle \langle \hat{c} \rangle)e^{-2i(\theta - \omega t)}].$$
(17)

$$\Delta Y^{2} = 2\kappa (2\langle \hat{c}^{\dagger} \hat{c} \rangle - 2\langle \hat{c}^{\dagger} \rangle \langle \hat{c} \rangle + 1) - \operatorname{Re}[4\kappa (\langle \hat{c} \hat{c} \rangle - \langle \hat{c} \rangle \langle \hat{c} \rangle)e^{-2i(\theta - \omega t)}].$$
(18)

Note that the factor of 2κ originates from the input-output relation in Eq. (11). In both Eqs. (17) and (18), the first term on the right-hand side indicates the incoherence of output field and the last term determines the squeezing of the output field. If the condition $\Delta X^2/2\kappa < 1$ or $\Delta Y^2/2\kappa < 1$ is fulfilled, the emitted light becomes quadrature-squeezed light.

III. RESULTS

In this section, we first analyze the third-order nonlinear optical response of cavity polaritons for different system sizes and show that there exists an optimal size of the exciton system where the nonlinear strength is maximum. Finally, we evaluate the light squeezing around the optimal size of the exciton system, in terms of the dependence of the squeezing on the incident light intensity.

Figure 3 shows the spectra of $|\xi_{out}^{(3)}|^2$ as a function of *N*. The pump energy is tuned to the energy of LPB, ω_{LPB} . For a system having no biexciton [Fig. 3(a)], strong nonlinearity is obtained only for small values of *N*. $|\xi_{out}^{(3)}|^2$ has its peak



FIG. 3. $|\xi^{(3)}|^2$ as a function of N for (a) a system having no biexciton and (b) a system having a stable biexciton. The parameters are $\kappa = 0.004$ eV, corresponding to a quality factor of Q = 400. The other parameters are the same as those used in Fig. 2.

at N = 6 and $\omega_{\text{probe}} \approx \omega_c - 0.004$ eV. As N increases, the nonlinear strength gradually decreases owing to the decrease in the unharmonicity originating from the discrete exciton states.

180

150

120

90

60

30

0

180

150

120

90

60

30

0

180

150

120

90

30

0

0

© 60

(_) θ

(_) θ

For a system with a stable biexciton [Fig. 3(b)], however, a strong nonlinearity is obtained widely for large values of N. $|\xi_{out}^{(3)}|^2$ has its peak at N = 60 and $\omega_{probe} \approx \omega_c - 0.015$ eV. Compared with Figs. 2(b) and 2(c), the value of N maximizing $|\xi_{out}^{(3)}|^2$ corresponds to that of N at which the biexciton states and the LPB [dashed line in Fig. 2(c)] intersect each other. Therefore an effective biexciton-polariton coupled state can be formed around the intersection point.⁷

The results in Fig. 3 imply that there exists an optimal size of the exciton system for a set of given parameters, for which the nonlinear strength becomes maximal: N = 6 for the system having no biexciton and N = 60 for that having a stable biexciton. Strong squeezing could be expected around these optimal sizes.

Figure 4 shows $\Delta X^2/2\kappa$ as a function of $|\xi_{in}|^2/2\kappa$ and θ at N = 2,6, and 10 for a system having no biexciton. In the absence of a biexciton, efficient squeezing is obtained in two regions: (i) $|\xi_{in}|^2/2\kappa \approx 0.015$ and $\theta \approx 160^\circ$ for N = 2 [Fig. 4(a)] and N = 10 [Fig. 4(c)], and (ii) $|\xi_{in}|^2/2\kappa \approx 0.004$ and $\theta \approx 30^\circ$ for N = 6 [Fig. 4(b)]. In (i), squeezing originates from saturation because it requires high intensity and can be observed for a large value of N. Meanwhile, the squeezing achieved at low intensity in (ii) is due to the exciton-exciton interaction, where the third-order nonlinearity is maximum, as shown in Fig. 3(a). Intriguingly, the squeezing remains almost unchanged with N (approximately 1% squeezing) though the third-order nonlinearity has its peak at N = 6.



FIG. 4. $\Delta X^2/2\kappa$ as a function of $|\xi_{\rm in}|^2/2\kappa$ and θ for a system with no biexciton. (a) N = 2, (b) N = 6, and (c) N = 10. $\omega = \omega_{\rm pump} = \omega_{\rm probe} = \omega_{\rm LPB}$.



FIG. 5. $\Delta X^2/2\kappa$ as a function of $|\xi_{\rm in}|^2/2\kappa$ and θ for a system with a stable biexciton. (a) N = 50, (b) N = 60, and (c) N = 70. $\omega = \omega_{\rm pump} = \omega_{\rm probe} = \omega_{\rm LPB}$.

Figure 5 shows $\Delta X^2/2\kappa$ as a function of $|\xi_{in}|^2/2\kappa$ and θ at N = 50,60, and 70 for a system having a stable biexciton. In the presence of a biexciton, efficient squeezing is obtained only in one region: $|\xi_{in}|^2/2\kappa \approx 0.004$ and $\theta \approx 0^\circ$. The squeezing becomes five times higher than that achieved in a system having no biexciton (approximately 5% squeezing). This is due to the interaction between the biexciton and cavity polaritons, and maximum squeezing can be achieved in this region as long as an effective biexciton-polariton coupled state is formed. Thus we can enhance light squeezing by utilizing the strong nonlinearity of a biexciton effectively coupled to cavity polaritons. It is simple to achieve efficient squeezing because a strong nonlinearity is obtained widely for large values of *N*, making the accurate control of *N* unnecessary.

IV. CONCLUSION AND DISCUSSION

In conclusion, we have theoretically investigated light squeezing via an interaction between a biexciton and cavity polaritons. We have analyzed in detail the nonlinear optical response of a biexciton-polariton coupled state, in terms of the dependence of the squeezing on the thickness of the quantum well, and evaluated the light squeezing by directly calculating the variance of quantum noise in an output field, in terms of dependence of the squeezing on the incident light intensity. We have shown that the squeezing utilizing the biexciton-polariton coupled state is five times higher than that achieved in a system having no biexciton. Such enhanced squeezing is achieved by controlling polariton level structure.

Throughout this work, we have considered a CuClquantum-well microcavity in which a biexciton can stably exist. If we consider a more general semiconductor system, such as a GaAS-quantum-well system, we would need to adopt a Wannier-like exciton model, and a calculation based on the dynamics-controlled truncation method¹² would be required. Further, a more exact fermionic treatment of excitons would then be necessary. In fact, a recent theoretical work has shown that fermionic statistics are related to the generation of squeezed light.¹³ In this study, fermionic property is artificially introduced by prohibiting the presence of two excitons at the same site, corresponding to the Pauli exclusion principle. However, it would be interesting to use microscopic theories of excitons starting with electrons and holes, properly including spin degree of freedom, Coulomb interaction between excitons, and biexciton formation. These factors might lead to further enhancement of the light squeezing. In addition, we have to take into account radiative and nonradiative decays of excitons because the Rabi slitting and biexciton binding energy become smaller than those of CuCl-cavity systems, and hence the effect of exciton damping on the light squeezing cannot be ignored. These are topics of our future studies.

Recently, a similar scheme using Feshbach resonance has been proposed,¹⁴ where the Rabi splitting formed by strong coupling of a biexciton and cavity polaritons is exploited. If the Q factor of a CuCl cavity system can be increased, this scheme could be achieved directly in our system. It might be interesting to compare with the results of this work.

Finally, the size dependence of nonlinearity might be more drastically changed in higher dimensions of the exciton system, and therefore further analyses of higher dimensional systems would be interesting. We hope that the results in this study help to identify some of the practical requirements for a squeezedlight source utilizing a biexciton nonlinearity.

ACKNOWLEDGMENTS

This work was supported by a Grant-in-Aid for Scientific Research (C) (No. 21540321) from Japan Society for the Promotion of Science, and by a Grant-in-Aid for Young Scientists (B) (No. 21760043) from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

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⁹If we consider higher excitation and properly analyze the effect of polariton saturation on light squeezing, we have to consider higher polariton states, such as $|3p\rangle = \alpha_{xxc}|k,k';1\rangle + \alpha_{xxc}|k;2\rangle + \alpha_{ccc}|G;3\rangle$.

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