

Exciton-induced squeezed state of light in semiconductors

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In this paper a mechanism for the generation of a squeezed state of light is theoretically presented. Due to coupling with excitons and via exciton-exciton interaction, a light beam propagating within a finite-size semiconductor sample is shown to evolve into a squeezed state. Based on an effective bosonic Hamiltonian, which is obtained from the original fermionic one, a detailed derivation of the photon quadrature variances is presented and dependences of the photon squeezing on both the extrinsic and intrinsic parameters are analyzed.

I. INTRODUCTION

Heisenberg uncertainty relations prevent the simultaneous exact measurements of any two physical quantities, whose operators do not commute, such as momentum and coordinate, field phase and particle number, two quadratures of a field, etc. There exist different states in which physical quantities are observed with errors. In chaotic states the product of squared errors of any pair of such quantities is always greater than $\frac{1}{16}$. The coherent state proposed by Schrödinger right after the birth of quantum mechanics and developed in the early 1960s by Glauber, Klauder, Sudarshan, and others (see, e.g., Ref. 1) is the state in which each of the two above-mentioned quantities share an equal amount of noise so that their squared error product reaches the shot-noise limit, $\frac{1}{16}$. It had been considered the “best” state until Stoler² suggested and mathematically formulated a “hypothetical” state that would allow one to “squeeze” the uncertainty domain of either of the two above quantities to zero, or more acceptably, to carry out precise measurements beyond the shot-noise limit (see also Ref. 3). This important state was later called the squeezed state.⁴ However, it took 15 years before it was observed experimentally.^{5–9} Because the squeezed state provides the potential capability of circumventing the limitations set by quantum noise in such applications as high-precision interferometry, optical communication, ultrasensitive laser spectroscopy, atomic spectroscopy, and gravitational wave detection, its subject as a major theoretical, experimental, and practical activity in quantum optics has been firmly established and studied with growing attention. Recently, squeezed states have been extended to solitons,^{10–12} polaritons,¹³ phonons,^{14,15} excitons,¹⁶ biexcitons,¹⁷ and so on. Of particular interest has been the investigation of squeezing in the elements of SU(1) and SU(2) algebras.^{10,11} The subject is thus of importance not only in quantum optics but also in quantum field theory, in condensed-matter physics, and in other areas of physics.

From a theoretical point of view squeezed states result from nonlinearities. The physical mechanisms of their generation should therefore be sought in nonlinear interactions. Since in a vacuum light-light coupling is ab-

sent, squeezed states of light must be realized in a material medium where light may indirectly interact with light via its interactions with the elementary excitations of the medium. Schemes for such material media are two-level or multilevel Jaynes-Cummings models and a variety of nonlinear optical systems of different geometrical configurations where light-squeezed states are produced due to second^{18–22} or higher²³ harmonic generation, four-wave mixing,^{3,24–29} parametric amplification,^{22,30,31} optical bistability^{31,32–34} and collective resonance fluorescence.^{35–37} Fabricated optical-fiber media³⁸ can also squeeze light via a Kerr-type nonlinearity.^{39–41} In Ref. 42 a metal was used as such a medium, where initially “linear” photons interacting with the electron plasma may become “nonlinear” and evolve into squeezed states. The present paper addresses semiconductors, which exhibit resonance-enhanced optical nonlinearities in the spectral range close to the band gap. In this energetic range the semiconductor exciton plays a key role in bringing about a good deal of interesting and new phenomena. Light propagating within a semiconductor can resonantly be coupled with excitons to become polaritons.⁴³ Because of their excitonic content, polaritons are strongly coupled to each other inside the semiconductor. Making use of the polariton picture, we evaluate the quadrature variances of photons as functions of linear ($\propto g$) and nonlinear ($\propto f$ and l) light-matter interactions. Also, externally tunable parameters, such as the initially prepared state and the size of the semiconductor sample as well as the frequency detuning between the exciting light beam and the exciton, enter our formulas in a quite compact fashion. This enables us to easily analyze the optimal conditions for detecting squeezed photons behind the semiconductor sample. We apply our analytic results to real semiconductors with well specified characteristics and perform necessary graphical illustrations which show the possibility of generating photon squeezed states through our exciton-induced mechanism. This squeezing mechanism seems to be among candidates for semiconductor operational devices.

Besides the introduction and conclusion sections, (Secs. I and V), this paper consists of three more sections. In Sec. II an effective bosonic Hamiltonian is obtained from

the original fermionic one. Section III is devoted to the photon quadrature variance evaluation. Section IV analyzes the dependence of the photon squeezing on the relevant parameters. There is also an Appendix which presents the detailed derivation of the main formula in Sec. III.

The system of units used throughout this paper is that with $\hbar=c=1$ where \hbar and c are, respectively, Planck's constant and the speed of light in vacuum.

$$H = \sum_{\mathbf{k}} \left[\omega(k)c_{\mathbf{k}}^{\dagger}c_{\mathbf{k}} + \varepsilon_e(k)e_{\mathbf{k}}^{\dagger}e_{\mathbf{k}} + \varepsilon_h(k)h_{\mathbf{k}}^{\dagger}h_{\mathbf{k}} + w(k) \sum_{\mathbf{p}} (c_{\mathbf{k}}^{\dagger}h_{\mathbf{p}}e_{\mathbf{k}-\mathbf{p}} + e_{\mathbf{k}-\mathbf{p}}^{\dagger}h_{\mathbf{p}}^{\dagger}c_{\mathbf{k}}) \right. \\ \left. + \frac{U(k)}{2V} \sum_{\mathbf{p}\mathbf{q}} (e_{\mathbf{p}}^{\dagger}e_{\mathbf{q}}^{\dagger}e_{\mathbf{q}-\mathbf{k}}e_{\mathbf{q}+\mathbf{k}} + h_{\mathbf{p}}^{\dagger}h_{\mathbf{q}}^{\dagger}h_{\mathbf{q}-\mathbf{k}}h_{\mathbf{q}+\mathbf{k}} - 2e_{\mathbf{p}}^{\dagger}h_{\mathbf{q}}^{\dagger}h_{\mathbf{q}-\mathbf{k}}e_{\mathbf{q}+\mathbf{k}}) \right], \quad (1)$$

where ω , ε_e , and ε_h are the photon, electron, and hole energies. For parabolic bands $\varepsilon_e(k)=E_g+k^2/(2m_e)$ and $\varepsilon_h(k)=k^2/(2m_h)$ with E_g the band gap and m_e (m_h) the effective mass of the electron (hole). $w(k)=-\sqrt{2\pi}(e/m)\Pi_{cv}[\varepsilon_0\omega(k)V]^{-1/2}$ with m (e) the free-electron mass (charge), ε_0 the static dielectric constant, Π_{cv} the interband matrix element of the momentum operator, and V the volume of the sample. $U(k)=4\pi e^2/(\varepsilon_0 k^2)$. c (c^{\dagger}), e (e^{\dagger}), and h (h^{\dagger}) denote second quantization operators of photons, electrons, and holes, respectively. At not too high optical excitations into the spectral range near but below the band edge the final-state electron-hole Coulomb interaction is of importance which likely makes electrons and holes bound in pairs called excitons. At finite density, excitons are not ideal bosons because of the residual Coulomb interactions among excitons as well as the effect due to the Pauli exclusion principle acting between the constituent particles belonging to different excitons. One of the possible approaches to the many-exciton system is to work out an effective Hamiltonian in which excitons appear as ideal bosons and at the same time the above-mentioned nonboson characteristics must be accounted for properly. To construct such a Hamiltonian we define one- and two-exciton state vectors in terms of both bosonic excitons and fermionic charged particles as below:

$$|ex; \nu\mathbf{k}\rangle = a_{\nu\mathbf{k}}^{\dagger}|0\rangle \\ = \frac{1}{\sqrt{V}} \sum_{\mathbf{p}} \varphi_{\nu}(\mathbf{k}-\beta\mathbf{p})e_{\mathbf{k}-\mathbf{p}}^{\dagger}h_{\mathbf{p}}^{\dagger}|0\rangle \quad (2)$$

$$H_{\text{eff}} = \sum_{\mathbf{k}} \left\{ \omega(k)c_{\mathbf{k}}^{\dagger}c_{\mathbf{k}} + \sum_{\nu} [E_{\nu}(k)a_{\nu\mathbf{k}}^{\dagger}a_{\nu\mathbf{k}} + g_{\nu}(k)(a_{\nu\mathbf{k}}^{\dagger}c_{\mathbf{k}} + c_{\mathbf{k}}^{\dagger}a_{\nu\mathbf{k}}) \right. \\ \left. + \frac{1}{V} \sum_{\mathbf{p}\mathbf{q}} \sum_{\nu\mu\xi\zeta} [f_{\nu\mu\xi\zeta}(\mathbf{k}, \mathbf{p}, \mathbf{q})a_{\nu\mathbf{p}}^{\dagger}a_{\mu\mathbf{q}}^{\dagger}a_{\xi\mathbf{p}-\mathbf{k}}a_{\zeta\mathbf{q}+\mathbf{k}} + l_{\nu\mu\xi}(\mathbf{k}, \mathbf{p}, \mathbf{q})a_{\nu\mathbf{p}}^{\dagger}a_{\mu\mathbf{q}}^{\dagger}a_{\xi\mathbf{p}-\mathbf{k}}c_{\mathbf{q}+\mathbf{k}} + \text{H. c.}] \right\}. \quad (7)$$

The exciton energy $E_{\nu}(k)$ and the effective interactions $g_{\nu}(k)$, $f_{\nu\mu\xi\zeta}(\mathbf{k}, \mathbf{p}, \mathbf{q})$, and $l_{\nu\mu\xi}(\mathbf{k}, \mathbf{p}, \mathbf{q})$ are determined from a hydrogenlike equation (8) and matrix elements (9)–(11) of H between the corresponding fermionic initial and final states

II. EFFECTIVE BOSONIC HAMILTONIAN

Consider for simplicity a two-band semiconductor with direct band gap and allowed interband transition. If the semiconductor is excited optically, there appear a number of conduction electrons which leave behind holes in the valence band. Treating the light inside the semiconductor as photons, the Hamiltonian of the resulting photon-electron-hole system has the form:

$$|ex, ex; \nu\mathbf{k}, \nu'\mathbf{k}'\rangle = P_{\nu\mathbf{k}, \nu'\mathbf{k}'} a_{\nu\mathbf{k}}^{\dagger} a_{\nu'\mathbf{k}'}^{\dagger} |0\rangle, \\ = \frac{P_{\nu\mathbf{k}, \nu'\mathbf{k}'}}{V} \sum_{\mathbf{p}\mathbf{p}'} \varphi_{\nu}(\mathbf{k}-\beta\mathbf{p})\varphi_{\nu'}(\mathbf{k}'-\beta\mathbf{p}') \\ \times e_{\mathbf{k}-\mathbf{p}}^{\dagger}h_{\mathbf{p}}^{\dagger}e_{\mathbf{k}'-\mathbf{p}'}^{\dagger}h_{\mathbf{p}'}^{\dagger}|0\rangle. \quad (3)$$

In (2) and (3), $a_{\nu\mathbf{k}}(a_{\nu\mathbf{k}}^{\dagger})$ denotes the bosonic operator of an exciton in a quantum state characterized by $\nu\mathbf{k}$ with $\nu=nlm$, φ_{ν} is the wave function describing the electron-hole relative motion in the momentum space, $\beta=m_h/M$ with $M=m_e+m_h$ and $P_{\nu\mathbf{k}, \nu'\mathbf{k}'}=1+(1/\sqrt{2}-1)\delta_{\nu\nu'}\delta_{\mathbf{k}\mathbf{k}'}$. For the one- and two-photon states we have, as usual,

$$|\gamma; \mathbf{k}\rangle = c_{\mathbf{k}}^{\dagger}|0\rangle, \quad (4)$$

$$|\gamma\gamma; \mathbf{k}, \mathbf{k}'\rangle = P_{\mathbf{k}\mathbf{k}'} c_{\mathbf{k}}^{\dagger} c_{\mathbf{k}'}^{\dagger}|0\rangle, \quad (5)$$

$P_{\mathbf{k}\mathbf{k}'}=1+(1/\sqrt{2}-1)\delta_{\mathbf{k}\mathbf{k}'}$, while the state containing one exciton and one photon is

$$|ex, \gamma; \nu\mathbf{k}, \mathbf{k}'\rangle = a_{\nu\mathbf{k}}^{\dagger} c_{\mathbf{k}'}^{\dagger}|0\rangle \\ = \frac{1}{\sqrt{V}} \sum_{\mathbf{p}} \varphi_{\nu}(\mathbf{k}-\beta\mathbf{p})e_{\mathbf{k}-\mathbf{p}}^{\dagger}h_{\mathbf{p}}^{\dagger}c_{\mathbf{k}'}^{\dagger}|0\rangle. \quad (6)$$

The effective Hamiltonian of (1), H_{eff} , can now be formulated through bosonic exciton operators a and a^{\dagger} as

$$\{\varepsilon_e[(1-\beta)\mathbf{p}+\mathbf{q}]+\varepsilon_h(\beta\mathbf{p}+\mathbf{q})\}\varphi_v(\mathbf{q})-\frac{1}{V}\sum_{\mathbf{q}'}U(\mathbf{q}-\mathbf{q}')\varphi_v(\mathbf{q}')=E_v(p)\varphi_v(\mathbf{q}), \quad (8)$$

$$g_v(k)=\langle v\mathbf{k};ex|H|\gamma;\mathbf{k}\rangle, \quad (9)$$

$$f_{\nu\mu\xi\xi}(\mathbf{k},\mathbf{p},\mathbf{q})=\langle \nu\mathbf{p},\mu\mathbf{q};ex,ex|H|ex,ex;\xi\mathbf{p}-\mathbf{k},\xi\mathbf{q}+\mathbf{k}\rangle, \quad (10)$$

$$l_{\nu\mu\xi}(\mathbf{k},\mathbf{p},\mathbf{q})=\langle \nu\mathbf{p},\mu\mathbf{q};ex,ex|H|ex,\gamma;\xi\mathbf{p}-\mathbf{k},\mathbf{q}+\mathbf{k}\rangle. \quad (11)$$

The solution of (8) yields $\varphi_v(\mathbf{q})$ in the form of hydrogenlike wave functions, and $E_v(k)=E_g-Ry/\nu^2+k^2/(2M)$ for $\nu=1,2,3,\dots$, and $E_\nu(k)=E_g+\nu^2/(2m_{eh})+k^2/(2M)$ for ν being a continuous number; Ry is the exciton Rydberg and $m_{eh}=m_e m_h/M$. Substituting (1) and the RHS of (2)–(6) into (9)–(11), we get

$$g_v(k)=\frac{w(k)}{V}\sum_{\mathbf{p}}\varphi_v(\mathbf{p}), \quad (12)$$

$$f_{\nu\mu\xi\xi}(\mathbf{k},\mathbf{p},\mathbf{q})=\sum_{i=1}^6 f_{\nu\mu\xi\xi}^{(i)}(\mathbf{k},\mathbf{p},\mathbf{q}), \quad (13)$$

where $f^{(i)}$ for $i=1,2,3$ depends only on k and describes the direct Coulomb interactions between the constituent particles belonging to different excitons, while $f^{(i)}$ for $i=4,5,6$ depends also on the difference $\mathbf{Q}=\mathbf{p}-\mathbf{q}$ and describes exchange interactions caused by the Pauli exclusion principle,

$$f_{\nu\mu\xi\xi}^{(1)}(k)=\frac{U(k)}{V}\sum_{\mathbf{k}_1}\varphi_\nu(\mathbf{k}_1-\beta\mathbf{k})\varphi_\xi^*(\mathbf{k}_1)\frac{1}{V}\sum_{\mathbf{k}_2}\varphi_\mu(\mathbf{k}_2-\beta\mathbf{k})\varphi_\xi^*(\mathbf{k}_2), \quad (14)$$

$$f_{\nu\mu\xi\xi}^{(2)}(k)=\frac{U(k)}{V}\sum_{\mathbf{k}_1}\varphi_\nu(\mathbf{k}_1+\mathbf{k}-\beta\mathbf{k})\varphi_\xi^*(\mathbf{k}_1)\frac{1}{V}\sum_{\mathbf{k}_2}\varphi_\mu(\mathbf{k}_2-\mathbf{k}+\beta\mathbf{k})\varphi_\xi^*(\mathbf{k}_2), \quad (15)$$

$$f_{\nu\mu\xi\xi}^{(3)}(k)=-\frac{2U(k)}{V}\sum_{\mathbf{k}_1}\varphi_\nu(\mathbf{k}_1-\beta\mathbf{k})\varphi_\xi^*(\mathbf{k}_1)\frac{1}{V}\sum_{\mathbf{k}_2}\varphi_\mu(\mathbf{k}_2-\mathbf{k}+\beta\mathbf{k})\varphi_\xi^*(\mathbf{k}_2), \quad (16)$$

$$f_{\nu\mu\xi\xi}^{(4)}(\mathbf{Q},\mathbf{k})=-\frac{1}{V^2}\sum_{\mathbf{k}_1\mathbf{k}_2}U(k_1)\varphi_\nu(\mathbf{k}_2-\mathbf{k}_1+\mathbf{k}-\beta\mathbf{k}) \\ \times\varphi_\mu(\mathbf{k}_2+\beta\mathbf{Q}+\beta\mathbf{k})\varphi_\xi^*(\mathbf{k}_2-\mathbf{k}_1+\beta\mathbf{Q}+\mathbf{k})\varphi_\xi^*(\mathbf{k}_2), \quad (17)$$

$$f_{\nu\mu\xi\xi}^{(5)}(\mathbf{Q},\mathbf{k})=-\frac{1}{V^2}\sum_{\mathbf{k}_1\mathbf{k}_2}U(k_1)\varphi_\nu(\mathbf{k}_2-\mathbf{k}_1-\beta\mathbf{k}) \\ \times\varphi_\mu(\mathbf{k}_2-\mathbf{k}-\mathbf{Q}+\beta\mathbf{Q}+\beta\mathbf{k})\varphi_\xi^*(\mathbf{k}_2-\mathbf{k}_1-\mathbf{Q}+\beta\mathbf{Q}+\mathbf{k})\varphi_\xi^*(\mathbf{k}_2), \quad (18)$$

$$f_{\nu\mu\xi\xi}^{(6)}(\mathbf{Q},\mathbf{k})=\frac{2}{V^2}\sum_{\mathbf{k}_1\mathbf{k}_2}U(k_1)\varphi_\nu(\mathbf{k}_2-\mathbf{k}_1-\beta\mathbf{k}) \\ \times\varphi_\mu(\mathbf{k}_2-\mathbf{Q}+\beta\mathbf{Q}-\mathbf{k}+\beta\mathbf{k})\varphi_\xi^*(\mathbf{k}_2-\mathbf{Q}+\beta\mathbf{Q}-\mathbf{k})\varphi_\xi^*(\mathbf{k}_2). \quad (19)$$

Note that our analytic expressions for the exciton-exciton interaction are, in fact, the same as those derived, e.g., in Refs. 44–49 by different methods. The influence of the particle spin was considered in Ref. 50 both analytically and numerically. Concerning the $l_{\nu\mu\xi}$ term, it describes a kind of collective response of the many-exciton system to the light: a photon can be absorbed (emitted) to generate (by destroying) an exciton with the assistance of another exciton coexisting in the medium with the former (see Fig. 1). These processes^{47,51} can occur only due to the exchange effect between the fermionic constituent particles of two excitons simultaneously participating in one and the same event with a photon. This kind of process allows excitons with many angular-momentum combinations (not only S ones) to interact with light. Their expressions read as

$$l_{\nu\mu\xi}(\mathbf{k},\mathbf{p},\mathbf{q})=-\frac{w(\mathbf{k}+\mathbf{q})}{V}\sum_{\mathbf{k}_1}\varphi_\nu(\mathbf{p}-\beta\mathbf{k}_1)\{\varphi_\mu[\mathbf{q}+\beta(\mathbf{p}-\mathbf{k}-\mathbf{k}_1-\mathbf{q})] \\ \times\varphi_\xi^*(\mathbf{p}-\mathbf{k}-\beta\mathbf{k}_1+\varphi_\mu[\mathbf{q}-\beta(\mathbf{k}_1-\mathbf{k})]\varphi_\xi^*[\mathbf{p}-\mathbf{k}-\beta(\mathbf{k}_1-\mathbf{k})]\}. \quad (20)$$

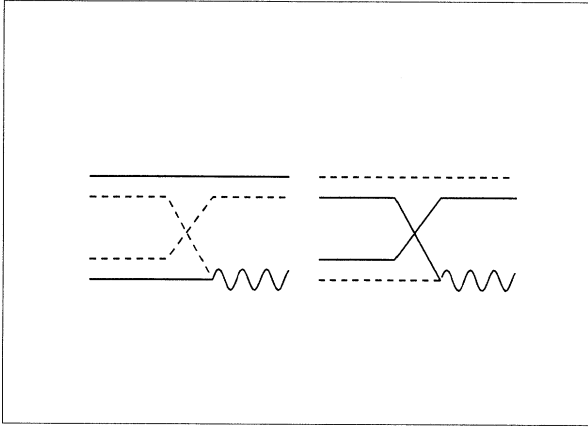


FIG. 1. Feynman diagrams showing the exciton-assisted exciton-photon transition, i.e., the l interaction in (24). Electron (hole) lines are solid (dashed). A solid line going in parallel with a dashed line represents an exciton. A crossing of solid (dashed) lines represents an exchange of two electrons (holes) belonging to two different excitons. Wavy lines are for photons. No arrows are indicated: each diagram can be read both from left to right (photon emission) and from right to left (photon absorption).

To further simplify the model Hamiltonian let us consider only one mode of photons with wave-vector \mathbf{k} and frequency ω very close to the lowest $\nu=1S$ exciton energy level. Since the $1S$ exciton has the strongest oscillator strength and is most actively coupled with the photon, we are interested only in these excitons which have the same momentum \mathbf{k} . Because of the smallness of the photon wave vector, k can then be put equal to zero and we do not write it any more in what follows. If only $\mathbf{k}=0$, $\nu=1S$ excitons exist, then (12), (14)–(19), and (20) can analytically be integrated to give⁵²

$$g \equiv g_{1S}(0) = -E \left[\frac{\epsilon_0 \Delta_{LT}}{2\omega} \right]^{1/2}, \quad (21)$$

$$f \equiv f_{1S1S1S}(0,0,0) = \frac{26\pi}{3} R y r_x^3, \quad (22)$$

$$l \equiv l_{1S1S1S}(0,0,0) = -7\pi g r_x^3. \quad (23)$$

In (21)–(23) $E = E_{1S}(0)$, Δ_{LT} is the exciton longitudinal-transverse splitting, and r_x is the exciton Bohr radius. The simplified effective Hamiltonian now becomes

$$H_{\text{eff}} = \omega c^+ c + E a^+ a + g(a^+ c + c^+ a) + \frac{f}{V} a^+ a^+ a a + \frac{l}{V} (a^+ a^+ a c + c^+ a^+ a a). \quad (24)$$

In (24) we put the dependence on the sample volume in clear evidence through V that, in fact, is the volume of a finite-size sample over which the coherent excitons and photons exist. We shall show that this “coherence volume” is essential in the squeezing phenomenon to be considered here. Such a feature was omitted in Ref. 53. Also, Ref. 53 did not yet take into account the so-called

exciton-assisted exciton-photon transition proportional to l in (24) and described by Feynman diagrams in Fig. 1.

III. PHOTON QUADRATURE VARIANCE EVALUATION

To study photon squeezing one needs to analyze the time-varying behavior of the normally ordered quadrature variances denoted by

$$X(t) = \langle N[\Delta x(t)]^2 \rangle, \quad P(t) = \langle N[\Delta p(t)]^2 \rangle \quad (25)$$

where the average $\langle \rangle$ is understood in the quantum sense, the N symbol normally orders all the operators standing after it, $\Delta x = x - \langle x \rangle$, $\Delta p = p - \langle p \rangle$, and

$$x(t) = \frac{1}{2}[c^+(t) + c(t)], \quad p(t) = \frac{i}{2}[c^+(t) - c(t)]. \quad (26)$$

As is well known, in a squeezed state one of the two variances is negative while the other is positive. The more the state is squeezed the greater is the absolute value of the negative variance. To see whether a variance may be negative during the time-evolution we need solving (24) for $c(t)$ and $c^+(t)$. For this aim, we perform the following Bogolubov transformations into new operators $\alpha_\nu(t)$ and $\alpha_\nu^+(t)$ with $\nu=1$ or 2 :

$$\begin{aligned} \alpha_\nu(t) &= u_\nu c(t) + v_\nu a(t), \\ \alpha_\nu^+(t) &= u_\nu c^+(t) + v_\nu a^+(t). \end{aligned} \quad (27)$$

The as yet unknown real functions u_ν and v_ν will then be determined so that the new operators $\alpha_\nu(t)$, $\alpha_\nu^+(t)$ will be bosonic and the transformed quadratic part of (24) will be diagonal. For the former, u_ν and v_ν should obey the orthonormalization condition

$$u_\nu u_\mu + v_\nu v_\mu = \delta_{\nu\mu}. \quad (28)$$

In particular, the normalization condition is

$$u_\nu^2 + v_\nu^2 = 1. \quad (29)$$

For the quadratic part of (24) to be transformed into a diagonal form, u_ν and v_ν should be related to each other as

$$v_\nu = \frac{g u_\nu}{\Omega_\nu - E}, \quad (30)$$

where Ω_ν is given by

$$\Omega_\nu = \frac{1}{2} \{ \omega + E + (-1)^\nu [(\omega - E)^2 + 4g^2]^{1/2} \}. \quad (31)$$

Making use of (31) we can establish an useful expression as follows:

$$(\Omega_1 - E)(\Omega_2 - E) = -g^2. \quad (32)$$

Combining (29) and (30) we get

$$u_\nu^2 = \frac{(\Omega_\nu - E)^2}{(\Omega_\nu - E)^2 + g^2}. \quad (33)$$

Now, the orthogonality condition

$$u_1 u_2 + v_1 v_2 = 0 \quad (34)$$

is easily provable with the aid of (30) and (32). Using (29), (30), (32), and (33), we can also check the following relations:

$$\sum_{\nu} u_{\nu}^2 = \sum_{\nu} v_{\nu}^2 = 1 \text{ and } \sum_{\nu} u_{\nu} v_{\nu} = 0. \quad (35)$$

Note that so far we have used only u_{ν}^2 in (33) but not u_{ν} . There is an uncertainty regarding the sign of u_{ν} . This uncertainty, however, does not affect the conditions (28) and (35). Therefore we can choose the sign of u_{ν} so that it is positive, i.e.,

$$u_{\nu} = \frac{|\Omega_{\nu} - E|}{[(\Omega_{\nu} - E)^2 + g^2]^{1/2}} = \frac{1}{\left[1 + \frac{g^2}{(\Omega_{\nu} - E)^2}\right]^{1/2}}. \quad (36)$$

The original operators $a(t)$ and $c(t)$ can be expressed in terms of the new ones, $\alpha_{\nu}(t)$, with the help of (27) and (35)

$$a(t) = \sum_{\nu} v_{\nu} \alpha_{\nu}(t), \quad a^{\dagger}(t) = \sum_{\nu} v_{\nu} \alpha_{\nu}^{\dagger}(t), \quad (37)$$

$$c(t) = \sum_{\nu} u_{\nu} \alpha_{\nu}(t), \quad c^{\dagger}(t) = \sum_{\nu} u_{\nu} \alpha_{\nu}^{\dagger}(t). \quad (38)$$

Relations (37) and (38) allow us to rewrite the whole H_{eff} in (24) in the form

$$H_{\text{eff}} = \sum_{\nu} \Omega_{\nu} \alpha_{\nu}^{\dagger} \alpha_{\nu} + \frac{1}{V} \sum_{\nu \mu \xi \zeta} [f v_{\nu} v_{\mu} v_{\xi} v_{\zeta} + l(v_{\nu} v_{\mu} v_{\xi} u_{\zeta} + u_{\nu} v_{\mu} v_{\xi} v_{\zeta})] \times \alpha_{\nu}^{\dagger} \alpha_{\mu}^{\dagger} \alpha_{\xi} \alpha_{\zeta}, \quad (39)$$

where each of ν , μ , ξ , and ζ takes a value of either 1 or 2. The new operators α_{ν} , α_{ν}^{\dagger} are interpreted as operators of a new two-branch quasiparticle called the polariton.⁴³ $\nu=1$ (2) corresponds to polaritons of the lower (upper) branch. The second quadruple sum in (39) describes scatterings among the polaritons. There are two scattering channels. One channel conserves the polariton numbers of each branch. It corresponds to the terms $\propto \alpha_{\nu}^{\dagger} \alpha_{\mu}^{\dagger} \alpha_{\mu} \alpha_{\nu}$. All the remaining terms are responsible for the other scattering channel which changes the polar-

iton number of a branch. Since there is an energy gap between the two polariton branches [see (31)], the latter scattering channel is energetically less probable and, in what follows, we will keep only the terms corresponding to the first scattering channel. This is referred to as secular approximation.⁵⁴ Let us now open (39) within the secular approximation to have the following form:

$$H_{\text{eff}} = \Omega_1 \alpha_1^{\dagger} \alpha_1 + \Omega_2 \alpha_2^{\dagger} \alpha_2 + \frac{1}{V} [F_{11} \alpha_1^{\dagger} \alpha_1^{\dagger} \alpha_1 \alpha_1 + F_{12} \alpha_1^{\dagger} \alpha_2^{\dagger} \alpha_2 \alpha_1 + F_{21} \alpha_2^{\dagger} \alpha_1^{\dagger} \alpha_1 \alpha_2 + F_{22} \alpha_2^{\dagger} \alpha_2^{\dagger} \alpha_2 \alpha_2], \quad (40)$$

where

$$F_{11} = v_1^3 (f v_1 + 2l u_1), \quad (41)$$

$$F_{12} = v_1 v_2^2 (f v_1 + 2l u_1), \quad (42)$$

$$F_{21} = v_1^2 v_2 (f v_2 + 2l u_2), \quad (43)$$

$$F_{22} = v_2^3 (f v_2 + 2l u_2). \quad (44)$$

Note that the form of (40) is reminiscent of a model Hamiltonian for two modes of light coupled to each other via a lossless nonlinear medium.⁵⁵ However, here in (40) the coupling constants $F_{\nu\mu}$ are specific. They depend explicitly on semiconductor parameters through f and l as well as on the photon frequency detuning, $\omega - E$, and on the direct light-matter interaction g through the Bogolubov transformation coefficients u_{ν} and v_{ν} . Using (40) we set up the Heisenberg equation of motion for the number operators $\alpha_{\nu}^{\dagger}(t) \alpha_{\nu}(t)$ and see, indeed,

$$\frac{d}{dt} [\alpha_{\nu}^{\dagger}(t) \alpha_{\nu}(t)] = i [H_{\text{eff}}, \alpha_{\nu}^{\dagger}(t) \alpha_{\nu}(t)] = 0, \quad (45)$$

i.e., $\alpha_{\nu}^{\dagger}(t) \alpha_{\nu}(t) = \alpha_{\nu}^{\dagger}(0) \alpha_{\nu}(0) = \text{const}$. The equations of motion for $\alpha_{\nu}(t)$,

$$\begin{aligned} \frac{d}{dt} \alpha_{\nu}(t) &= i [H_{\text{eff}}, \alpha_{\nu}(t)] \\ &= -i \left[\Omega_{\nu} + \frac{1}{V} \sum_{\mu} (F_{\nu\mu} + F_{\mu\nu}) \alpha_{\mu}^{\dagger}(t) \alpha_{\mu}(t) \right] \alpha_{\nu}(t), \end{aligned} \quad (46)$$

are then easy to solve because the products $\alpha_{\mu}^{\dagger}(t) \alpha_{\mu}(t)$ in the square brackets are constant. The solutions read as

$$\alpha_{\nu}(t) = \exp \left\{ -i \left[\Omega_{\nu} + \frac{1}{V} \sum_{\mu} (F_{\nu\mu} + F_{\mu\nu}) \alpha_{\mu}^{\dagger}(0) \alpha_{\mu}(0) \right] t \right\} \alpha_{\nu}(0), \quad (47)$$

$$\alpha_{\nu}^{\dagger}(t) = \alpha_{\nu}^{\dagger}(0) \exp \left\{ i \left[\Omega_{\nu} + \frac{1}{V} \sum_{\mu} (F_{\nu\mu} + F_{\mu\nu}) \alpha_{\mu}^{\dagger}(0) \alpha_{\mu}(0) \right] t \right\}. \quad (48)$$

Substituting (47) and (48) into (38) gives immediately the time dependence of the photon operators. Nevertheless, $c(t)$ and $c^{\dagger}(t)$ obtained in such a way still contain in themselves the products $\alpha_{\mu}^{\dagger}(0) \alpha_{\mu}(0)$. Although these products are time independent, the evaluation of $X(t)$ and $P(t)$ remains nontrivial and, hence, will be done now in detail. Suppose at $t=0$ the excitons and photons are

in their coherent states characterized, respectively, by complex numbers z_a and z_c ,

$$z_a = \sqrt{N_a} \exp(i\theta_a), \quad z_c = \sqrt{N_c} \exp(i\theta_c), \quad (49)$$

where N_a , θ_a and N_c , θ_c are any real numbers. N_a and N_c represent average exciton and photon numbers, re-

spectively. The initial state of the coherent exciton-photon system can be written as

$$|z_a, z_c\rangle = D_a(z_a)D_c(z_c)|0\rangle, \quad (50)$$

where the displacement operator $D_b(z_b)$ has the following form for any bosonic operator b

$$D_b(z_b) = \exp(z_b b^\dagger - z_b^* b). \quad (51)$$

As seen from (38), (47), (48), and (26), $X(t)$ and $P(t)$ in (25) are fully determined by the operator behavior at $t=0$. Thus, we can write

$$\langle \rangle = \langle 0|D_c^\dagger(z_c)D_a^\dagger(z_a) \cdots D_a(z_a)D_c(z_c)|0\rangle. \quad (52)$$

Making use of (51) and (26) and of the fact that $\alpha_v, \alpha_v^\dagger$ are bosonic operators, we establish the following relationships:

$$D_a(z_a) = D_{\alpha_1}(v_1 z_a)D_{\alpha_2}(v_2 z_a), \quad (53)$$

$$D_c(z_c) = D_{\alpha_1}(u_1 z_c)D_{\alpha_2}(u_2 z_c). \quad (54)$$

With these at hand, we cast (52) into

$$\begin{aligned} \langle \rangle &= \langle 0|D_{\alpha_2}^\dagger(u_2 z_c)D_{\alpha_1}^\dagger(u_1 z_c)D_{\alpha_2}^\dagger(v_2 z_a)D_{\alpha_1}^\dagger(v_1 z_a) \\ &\quad \times \cdots D_{\alpha_1}(v_1 z_a)D_{\alpha_2}(v_2 z_a) \\ &\quad \times D_{\alpha_1}(u_1 z_c)D_{\alpha_2}(u_2 z_c)|0\rangle. \end{aligned} \quad (55)$$

For definiteness from now on we shall be interested in $P(t)$ only. In terms of photon operators $P(t)$ is

$$P(t) = \frac{1}{2}[\langle c^\dagger(t)c(t)\rangle - \text{Re}\langle c^2(t)\rangle - 2[\text{Im}\langle c(t)\rangle]^2]. \quad (56)$$

Putting (38) into (56) we can express $P(t)$ in terms of polariton operators as

$$\begin{aligned} P(t) &= \frac{1}{2}\{u_1^2[\langle \alpha_1^\dagger(t)\alpha_1(t)\rangle - \text{Re}\langle \alpha_1^2(t)\rangle - 2[\text{Im}\langle \alpha_1(t)\rangle]^2] \\ &\quad + 2u_1 u_2[\text{Re}\langle \alpha_2^\dagger(t)\alpha_1(t)\rangle - \text{Re}\langle \alpha_2(t)\alpha_1(t)\rangle - 2\text{Im}\langle \alpha_2(t)\rangle\text{Im}\langle \alpha_1(t)\rangle] \\ &\quad + u_2^2[\langle \alpha_2^\dagger(t)\alpha_2(t)\rangle - \text{Re}\langle \alpha_2^2(t)\rangle - 2[\text{Im}\langle \alpha_2(t)\rangle]^2]\}. \end{aligned} \quad (57)$$

The various averages entering (57) are understood in accordance with (55). Assuming for simplicity $\theta_a = \theta_c = 0$, we have obtained the final result which is formulated in the form

$$\begin{aligned} P(t) &= \frac{u_1^2 Q_1^2}{2}\{1 - \cos(r_{20}^{12} + x_{11}t)\exp(s_{20}^{12}) - 2\sin^2(r_{10}^{12})\exp(2s_{10}^{12})\} \\ &\quad + u_1 u_2 Q_1 Q_2\{\cos(r_{11}^{12})\exp(s_{11}^{12}) - \cos(r_{1-1}^{12} + x_{12}t)\exp(s_{1-1}^{12}) - 2\sin(r_{10}^{12})\sin(r_{10}^{21})\exp(s_{10}^{12} + s_{10}^{21})\} \\ &\quad + \frac{u_2^2 Q_2^2}{2}\{1 - \cos(r_{20}^{21} + x_{22}t)\exp(s_{20}^{21}) - 2\sin^2(r_{10}^{21})\exp(2s_{10}^{21})\}, \end{aligned} \quad (58)$$

where

$$\begin{aligned} Q_v &= u_v \sqrt{N_c} + v_v \sqrt{N_a}, \quad x_{\nu\mu} = (F_{\nu\mu} + F_{\mu\nu})/V, \\ r_{nm}^{\nu\mu} &= (n\Omega_\nu - m\Omega_\nu)t - Q_\nu^2 \sin[(mx_{\nu\mu} - nx_{\nu\nu})t] - Q_\mu^2 \sin[(mx_{\mu\mu} - nx_{\mu\nu})t], \\ s_{nm}^{\nu\mu} &= -2\left\{Q_\nu^2 \sin^2\left[(mx_{\nu\mu} - nx_{\nu\nu})\frac{t}{2}\right] + Q_\mu^2 \sin^2\left[(mx_{\mu\mu} - nx_{\mu\nu})\frac{t}{2}\right]\right\}. \end{aligned} \quad (59)$$

Equation (58) is analytically exact within the secular approximation. Its detailed derivation will be given in the Appendix.

For $x_{\nu\mu}t \ll 1$, $s_{nm}^{\nu\mu}$ can be put equal to zero, while

$$r_{nm}^{\nu\mu} \rightarrow (np_{\nu\mu} - mp_{\mu\nu})t \quad (60)$$

with

$$p_{\nu\mu} = \Omega_\nu + Q_\nu^2 x_{\nu\nu} + Q_\mu^2 x_{\mu\nu}. \quad (61)$$

Then (58) becomes

$$\begin{aligned} P(t) &= u_1^2 Q_1^2 \sin\left[\left[2p_{12} + \frac{x_{11}}{2}\right]t\right] \sin\left[\frac{x_{11}}{2}t\right] \\ &\quad + 2u_1 u_2 Q_1 Q_2 \sin\left[\left[p_{12} + p_{21} + \frac{x_{12}}{2}\right]t\right] \sin\left[\frac{x_{12}}{2}t\right] + u_2^2 Q_2^2 \sin\left[\left[2p_{21} + \frac{x_{22}}{2}\right]t\right] \sin\left[\frac{x_{22}}{2}t\right]. \end{aligned} \quad (62)$$

IV. APPLICATION TO REAL SEMICONDUCTOR

Formula (58) for the photon P -quadrature variance displays several interesting features concerning the role of different kinds of interactions and the dependence on controllable parameters. Already from (40)–(44) we saw that the true interactions g , f , and l [see (24)] manifest themselves not explicitly separately but rather in combinations through the effective interactions $F_{\nu\mu}$. These $F_{\nu\mu}$ enter (58) through $x_{\nu\mu}$. Using the asymptotic expression (62) we can immediately conclude that no squeezing will occur if all of the $x_{\nu\mu}=0$, i.e., all of the $F_{\nu\mu}=0$, because then the second sinusoidal functions on each line of (62) exactly equal zero. Returning back for a moment to (41)–(44), we find out that all the $F_{\nu\mu}$ will vanish in either of the two following cases: (i) $f=l=0$ and (ii) all the $u_\nu=0$. The former is easy to understand since it means no nonlinearities. The latter takes place when $g=0$ [see Eq. (30)]. Squeezing again is absent because, physically, $g=0$ means no light-matter interaction. Thus, the necessary conditions for squeezing to occur are simultaneous nonvanishing of g and of at least one of the two f and l (not necessarily of both of them). These are intrinsic parameters, i.e., material-dependent ones. The extrinsic controllable parameters are the frequency detuning ($\omega-E$), the initial average numbers of excitons and photons N_a , N_c , and the volume V of the sample. To make visual the dependence on the extrinsic parameters we fix the intrinsic ones by choosing a concrete semiconductor, say, CdS. The parameters for CdS are $E_g=2.586$ eV, $E=2.553$ eV, $\Delta_{LT}=1$ meV, $\epsilon_0=8$ and $r_x=25.5$ Å. These values give $g\approx-101$ meV, $f\approx 1.5\times 10^{-17}$ meV cm³ and $l\approx 3.7\times 10^{-17}$ meV cm³. In Fig. 2 we draw P as a function of Et for $N_a=N_c=100$, $V/v_{ex}=4\times 10^3$, where v_{ex} is the volume of an exciton (for CdS $v_{ex}\approx 7\times 10^{-20}$ cm³), and different scaled detunings in terms of the ratio $\omega/E=0.9$, 0.95, and 1. We see P develops negative parts indicating the occurrence of the photon squeezing. The smaller the detuning, i.e., the nearer the ratio ω/E to 1, the deeper the negative peaks,

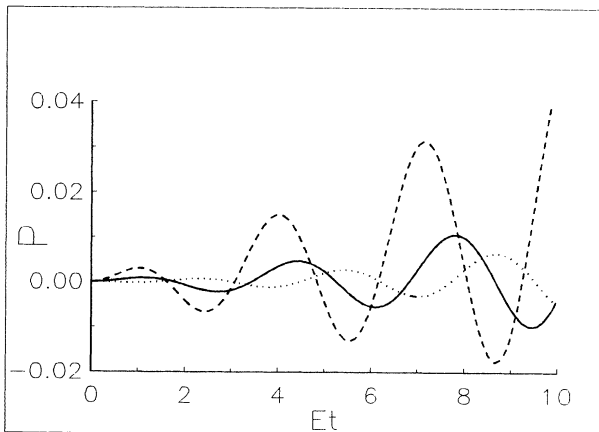


FIG. 2. P -quadrature variance of CdS vs scaled time Et . $N_a=N_c=100$, $V/v_{ex}=4\times 10^3$ and ω/E varies. The dotted (solid and dashed) curve corresponds to $\omega/E=0.9$ (0.95 and 1).

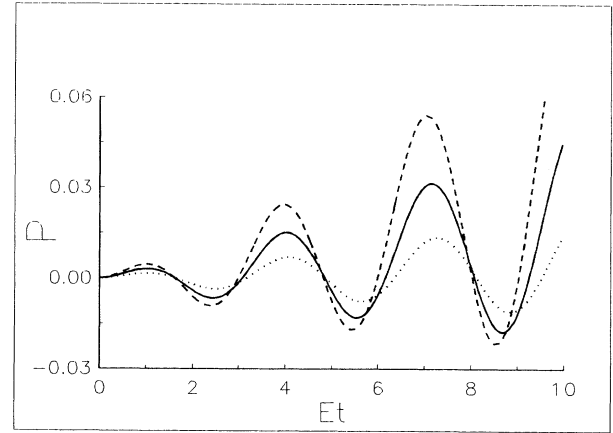


FIG. 3. Same as Fig. 1 but $\omega/E=1$, $V/v_{ex}=4\times 10^3$ and $N_a=N_c=50$, 100, and 150 corresponding to the dotted, solid, and dashed curve.

i.e., the better the squeezing. The perfect resonance detuning, $\omega/E=1$, causes the best squeezing. Figure 3 plots the same $P=P(Et)$ with fixed $\omega/E=1$, $V/v_{ex}=4\times 10^3$ and varying $N_a=N_c=50$, 100, and 150. Clearly seen that a better squeezing corresponds to a larger initial numbers of excitons and photons. Figure 4 shows the variation of squeezing in dependence on V ($V/v_{ex}=4\times 10^3$, 8×10^3 , and 2×10^4) when $\omega/E=1$ and $N_a=N_c=100$. One sees that smaller sample volume more favors the squeezing process. Qualitatively similar behaviors hold as well for GaAs whose parameters are $E_g=1.5$ eV, $E=1.495$ eV, $\Delta_{LT}=0.1$ meV, $\epsilon_0=12$ and $r_x=100$ Å which give $g\approx-30$ meV, $f\approx 1.4\times 10^{-16}$ meV cm³ and $l\approx 6.6\times 10^{-16}$ meV cm³.

To see the dependence on the intrinsic parameters, we shall proceed as follows. Namely, let us forget the concrete analytic expressions (21)–(23), and treat g , f , and l as varying parameters representing a change from one to another semiconductor. We then analyze the squeezing

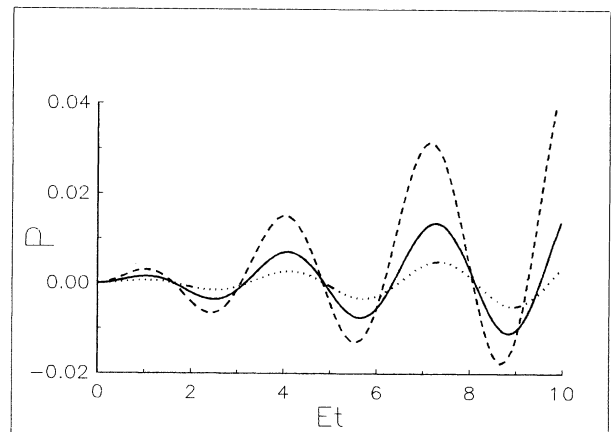


FIG. 4. Same as Fig. 1 but $\omega/E=1$, $N_a=N_c=100$ and V/v_{ex} varies. The dashed (solid and dotted) curve corresponds to $V/v_{ex}=4\times 10^3$ (8×10^3 and 2×10^4).

phenomenon as a function of such intrinsic variables. For that purpose it is convenient to deal with scaled quantities such as g/E , f/E , and l/E . From the above specific parameter values we have $g/E \approx -0.03696$ (-0.0200), $f/E \approx 5.8149 \times 10^{-21} \text{ cm}^3$ ($9.1014 \times 10^{-20} \text{ cm}^3$) and $l/E \approx 1.4426 \times 10^{-20} \text{ cm}^3$ ($4.6183 \times 10^{-19} \text{ cm}^3$) for CdS (GaAs). We therefore can let g/E vary from -0.06 to 0 , and F and L vary from 0 to 100 where $f/E = F \times 10^{-21} \text{ cm}^3$ and $l/E = L \times 10^{-20} \text{ cm}^3$. A three-dimensional (3D) plot in Fig. 5 illustrates an imaginary situation when $f=0$ and $l \neq 0$. For $L=0$ there is no squeezing. For increasing L the valleys become deeper meaning stronger squeezing. The importance of the interaction term proportional to l , the l interaction, is emphasized by the fact that squeezing can happen even in the case of $f=0$. Generally both the f and the l interactions must be included since they are of almost the same order of magnitude at least for CdS and GaAs as seen from their values above. It is worth noting at this moment that the f interaction results from a dipole-dipole-like nature of the exciton-exciton interaction. In low dimensions, though the exciton Rydberg increases, the exciton-exciton interaction decreases because of more "neutralization" of each exciton. On the other hand, the l interaction is due to the interaction between a photon and an electron-hole pair and to the Pauli exchange effect acting between the electrons (holes) of two excitons (see Fig. 1). This l interaction is not influenced by the exciton neutralization and is expected to be of more importance in low dimensions than the f interaction. Our consideration of the l interaction from the very onset is thus necessary. In Fig. 6 we analyze the dependence on the coupling constant g when $f/E = l/E = 10^{-19} \text{ cm}^3$, $V = 10^{-15} \text{ cm}^3$, $N_a = N_c = 100$ and $\omega/E = 0.95$. As analytically noticed before, here we see graphically that squeezing can occur only for $g \neq 0$. Figures 5 and 6 indi-

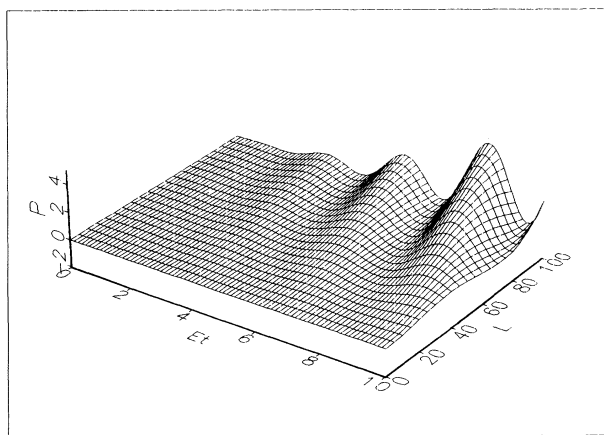


FIG. 5. P -quadrature variance vs scaled time Et and L (for L see text). The other parameters are taken as $g/E = -0.04$, $\omega/E = 1$, $N_a = N_c = 100$, $V = 10^{-15} \text{ cm}^3$, and $f = 0$.

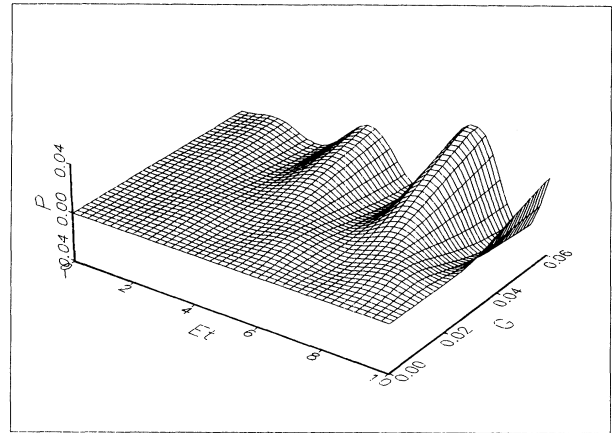


FIG. 6. P -quadrature variance vs Et and $G = -g/E$. The other parameters are taken as $\omega/E = 0.95$, $N_a = N_c = 100$, $V = 10^{-15} \text{ cm}^3$, and $l/E = f/E = 10^{-19} \text{ cm}^3$.

cate that for the squeezing process, semiconductors with larger interactions are desirable.

V. CONCLUSION

It is well known that excitonic nonlinearities have brought about a lot of interesting physical phenomena related to nonlinear coherent propagation of light in semiconductors such as nutation, Bose condensation, biexciton and multiexciton formation, bistability, self-induced transparency, photon echo, optical excitonic Stark effect, giant polariton, chaotic self-pulsation, etc. In this paper, we have shown one more effect caused by both the exciton-photon and exciton-exciton interactions which we call the exciton-induced squeezing of light. Among analyzed dependences on the extrinsic and intrinsic parameters, the dependence on the sample volume needs some more discussion. From the text (and see Fig. 4) we saw that the smaller the sample volume the better the squeezing. In plotting the figures we have chosen either $V/v_{\text{ex}} = 4 \times 10^3$, 8×10^3 , and 2×10^4 (for CdS) or $V = 10^{-15} \text{ cm}^3$. Such volumes are somewhat intermediate between bulk crystals and very large microcrystallites.^{51,56} One may think that further reducing the volume should lead to better squeezing. However, for smaller volume, the sample becomes a quantum box or a quantum dot and then the quantum size effect will need to be accounted for properly. Although formally for a microcrystallite one can still use an effective bosonic Hamiltonian⁵¹ like (24), the analytical expressions for E , g , f , and l are much different from those given by (8) and (21)–(23). Their calculation is even numerically a formidable task. Since nowadays semiconductor microcrystallite samples are in a good stage of fabrication, the above-proposed exciton-induced mechanism of generating photon squeezed states in them is worth to be considered. As a rule, nevertheless, this requires further more accurate investigations.

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APPENDIX

This appendix is to derive Eq. (58). For this we need the following operator identities:

$$D_b^+(z)bD_b(z)=b+z, \quad D_b^+(z)b^+D_b(z)=b^++z^*, \quad (\text{A1})$$

$$D_b(z_1)D_b(z_2)=D_b(z_1+z_2)e^{i\text{Im}(z_2^*z_1)}, \quad (\text{A2})$$

$$e^{\lambda b+b}D_b(z)=D_b(ze^\lambda)e^{\lambda b+b}, \quad (\text{A3})$$

$$\langle 0|D_b^+(z_1)D_b(z_2)|\rangle=\exp\left[-\frac{1}{2}|z_1-z_2|^2+i\text{Im}(z_1^*z_2)\right], \quad (\text{A4})$$

$$e^{\lambda b+b}=\sum_{n=0}^{\infty}\frac{(e^\lambda-1)^n}{n!}(b^+)^nb^n. \quad (\text{A5})$$

While the first four identities are properties of the displacement operators, the last one can be found, e.g., in Ref. 57. For convenience, we define two auxiliary functions $Q(z_1, z_2)$ and $S(z_1, z_2; \lambda)$ which are useful to compact the formulas

$$Q(z_1, z_2)=\frac{\langle 0|\dots bD_b(z_1)D_b(z_2)|0\rangle}{\langle 0|\dots D_b(z_1)D_b(z_2)|0\rangle}, \quad (\text{A6})$$

where “...” may be anything, and

$$S(z_1, z_2; \lambda)=\langle 0|D_b^+(z_2)D_b^+(z_1)\exp(\lambda b^+b)D_b(z_1)D_b(z_2)|0\rangle. \quad (\text{A7})$$

Double applying (A1) yields at once

$$Q(z_1, z_2)=z_1+z_2. \quad (\text{A8})$$

Concerning the function $S(z_1, z_2; \lambda)$, the use of (A2) gives

$$S(z_1, z_2; \lambda)=\langle 0|D_b^+(z_1+z_2)e^{\lambda b+b}D_b(z_1+z_2)|0\rangle. \quad (\text{A9})$$

Then the use of (A3) gives

$$S(z_1, z_2; \lambda)=\langle 0|D_b^+(z_1+z_2)D_b[(z_1+z_2)e^\lambda]e^{\lambda b+b}|0\rangle, \quad (\text{A10})$$

and, the use of (A5) gives

$$S(z_1, z_2; \lambda)=\sum_{n=0}^{\infty}\frac{(e^\lambda-1)^n}{n!}\langle 0|D_b^+(z_1+z_2)D_b[(z_1+z_2)e^\lambda](b^+)^nb^n|0\rangle. \quad (\text{A11})$$

In (A11) all the terms with $n=1, 2, 3, \dots$ vanish due to the action of b on $|0\rangle$. There remains only the term with $n=0$, i.e.,

$$S(z_1, z_2; \lambda)=\langle 0|D_b^+(z_1+z_2)D_b[(z_1+z_2)e^\lambda]|0\rangle. \quad (\text{A12})$$

Now the use of (A4) yields the final expression for $S(z_1, z_2; \lambda)$,

$$S(z_1, z_2; \lambda)=\exp[(e^\lambda-1)|z_1+z_2|^2]. \quad (\text{A13})$$

Having $Q(z_1, z_2)$ and $S(z_1, z_2; \lambda)$ at hand, it is easy to find by applying (47), (48), and (55) that

$$\begin{aligned} \langle \alpha_1(t) \rangle &= \exp(-i\Omega_1 t)Q(v_1 z_a, u_1 z_c) \\ &\quad \times S\left[u_1 z_c, v_1 z_a; -\frac{2i}{V}F_{11}t\right]S\left[u_2 z_c, v_2 z_a; -\frac{i}{V}(F_{12}+F_{21})t\right], \end{aligned} \quad (\text{A14})$$

$$\begin{aligned} \langle \alpha_1^2(t) \rangle &= \exp\left[-2i\left[\Omega_1 + \frac{F_{11}}{V}\right]t\right]Q^2(v_1 z_a, u_1 z_c) \\ &\quad \times S\left[u_1 z_c, v_1 z_a; -\frac{4i}{V}F_{11}t\right]S\left[u_2 z_c, v_2 z_a; -\frac{2i}{V}(F_{12}+F_{21})t\right], \end{aligned} \quad (\text{A15})$$

$$\langle \alpha_2(t) \alpha_1(t) \rangle = \exp \left[-i \left[\Omega_1 + \Omega_2 + \frac{F_{12} + F_{21}}{V} \right] t \right] Q(v_1 z_a, u_1 z_c) Q(v_2 z_a, u_2 z_c) \\ \times S \left[u_1 z_c, v_1 z_a; -\frac{i}{V} (2F_{11} + F_{12} + F_{21}) t \right] S \left[u_2 z_c, v_2 z_a; -\frac{i}{V} (2F_{22} + F_{12} + F_{21}) t \right], \quad (\text{A16})$$

$$\langle \alpha_2^+(t) \alpha_1(t) \rangle = \exp[-i(\Omega_1 - \Omega_2)t] Q^*(u_2 z_c, v_2 z_c) Q(u_1 z_c, v_1 z_a) \\ \times S \left[u_1 z_c, v_1 z_a; \frac{i}{V} (F_{12} + F_{21} - 2F_{11}) t \right] S \left[u_2 z_c, v_2 z_a; \frac{i}{V} (2F_{22} - F_{12} - F_{21}) t \right], \quad (\text{A17})$$

$$\langle \alpha_1^+(t) \alpha_1(t) \rangle = |Q(u_1 z_c, v_1 z_a)|^2. \quad (\text{A18})$$

As for $\langle \alpha_2(t) \rangle$, $\langle \alpha_2^2(t) \rangle$, and $\langle \alpha_2^+(t) \alpha_2(t) \rangle$, they are obtained, respectively, from (A14), (A15), and (A18) with an exchange of the subscript 1 (2) by 2 (1). Taking the necessary real and imaginary parts of (A14)–(A18) and putting them into (57), we have arrived at the formula (58) which is the main result of our paper.

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