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## On exciton squeezing in semiconductors

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**Abstract.** We consider an optically excited semiconductor as a coherent system of interacting excitons and photons. Within the boson formalism and secular approximation the analytical expressions for the exciton quadrature variances are rigorously derived. The analysis of these variances over the relevant parameter space shows that initially coherent excitons can evolve into squeezed states in a finite-size semiconductor, which must possess both linear and non-linear interactions.

### 1. Introduction

Squeezed states of light were theoretically proposed in 1970 [1]. Their first experimental observations in 1985 (see selected papers and reviews in [2]) made them a major subject in quantum optics, which has been unceasingly studied from both fundamental and practical points of view. This is due to the fact that in a squeezed state the uncertainty of one of the two quadrature components of the field is reduced below the coherence-state value [3], promising possibilities for precision measurements beyond the shot-noise limit. These open potential applications e.g. in interferometry, ultrasensitive laser spectroscopy, optical communication, detection of gravitational waves and so on. Apart from well known advantages in quantum optics, the concept of squeezed states possesses the common character that its analogies must also be found in other areas of physics such as atomic physics, quantum-field theory and condensed-matter physics. Squeezed states can therefore be produced not only for light but also for other kinds of quasiparticle existing in a material medium. Indeed, these have recently been considered for solitons [4–6], phonons [7, 8], polaritons [9–11], excitons [12, 13], biexcitons [14], etc. In [8] it was confirmed that the squeezed state of phonons gives a lower ground-state energy of the whole superconducting system than do the coherent and the displaced states. Polaritons as two-mode intrinsically squeezed states were studied in [9], where the experimental possibilities were also discussed for detecting the squeezed polariton structure. In [10] and [11] polariton squeezing in a small-volume crystal was shown to occur in both bunching and antibunching situations. [12] showed that the exciton in a photon–exciton system without non-linearities could periodically appear in a squeezed state if and only if the initial state of the system possesses a certain degree of squeezing. However, when non-linear interactions, for example, the exciton–exciton one, are included, squeezed excitons were demonstrated to be possible to generate by coherent light [13]: the system does not need to possess any initial degree of squeezing. Biexciton squeezing was also investigated in a photon–exciton system additionally pumped by a coherent light beam of finite intensity and of frequency resonant

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with the exciton–biexciton transition [14]. In the present paper, we develop further the topic of [12] and [13], taking into account an additional kind of non-linearity that can also cause exciton squeezing, and with more detailed examination of the dependence of exciton squeezing on all the relevant parameters in real semiconductors. The additional non-linearity just mentioned is the collective response of the many-exciton system to the light. Such a non-linearity is of the same order of magnitude as that of the exciton–exciton interaction in bulk CdS and GaAs, and furthermore, it proves to be of more significance in low-dimensional samples, e.g., in quantum wells and quantum dots. Its consideration from the very onset is thus necessary and will be done here. Furthermore, it is worth noting that the formulae for the exciton quadrature variances to be derived in this paper are analytically exact and much more compact than those reported in our previous work [13].

We organise our paper as follows. Section 2 deals with the model Hamiltonian, which is constructed within a boson formalism. This Hamiltonian is then applied in section 3 to derive the exciton quadrature variances. Section 4 analyses the time evolution of the variances over the parameter space. Finally, some relevant discussion is given in section 5.

We shall use the units with both the Planck constant  $\hbar$  and the velocity  $c$  of light in vacuum equal to unity.

## 2. Model Hamiltonian

Consider an optically excited two-simple-direct-band semiconductor, which can be modelled as a photon–electron–hole system: the photon creates or destroys the electron–hole pair, while the charged carriers interact with each other via Coulomb forces. Suppose the exciting light is spectroscopically close to the semiconductor band edge. Then the electron–hole pairs are likely to be formed in their bound states, called excitons. Since an exciton comprises an electron and a hole, it is obviously boson-like but not an ideal boson. The non-boson character can be dealt with by taking account of the exact commutation relations between non-bosonic exciton operators. This is the non-boson approach to the many-exciton system [15–22]. There is another approach that makes the best use of the boson-like nature of excitons to introduce a hypothetical boson space in which the exciton exact kinetics as well as the residual interactions between different electron–hole pairs may be described as effective interactions along the bosonic excitons. The effective bosonic Hamiltonian has been derived from the original photon–electron–hole one by several methods without [23–28] and with [29] spin effects taken into account. If interested in only one mode of photons with wave-vector  $k_0$  and frequency  $\omega$  very close to the lowest  $n = 1S$  exciton energy level  $E = E_{1S}$ , one can neglect (see e.g. [23] and [24]) the existence of all the excitons with  $k \neq k_0$  and  $n \neq 1S$ . Such an approximation is generally not well justified in real systems where scatterings of the ( $k = k_0, n = 1S$ ) excitons by phonons, impurities and also by each other will lead to the appearance of excitons with  $k \neq k_0$  and  $n \neq 1S$ . However, for the sake of simplicity, we shall in this paper confine ourselves only to a highly idealized situation in which we ignore all internal degrees of freedom connected with the relative electron–hole motion in an exciton as well as any scattering mechanisms that may destroy the coherency of the system. The idealized effective bosonic Hamiltonian for the exciton–photon system then has the following simple form:

$$H = \omega c^\dagger c + E a^\dagger a + g(a^\dagger c + c^\dagger a) + (f/V)a^\dagger a^\dagger a a + (l/V)(a^\dagger a^\dagger a c + c^\dagger a^\dagger a a) \quad (1)$$

where for brevity we have suppressed writing the indices  $k_0$  and  $n = 1S$ .  $c$  ( $c^\dagger$ ) and  $a$  ( $a^\dagger$ ) stand for the annihilation (creation) operators of the single-mode photon and the

lowest-energy exciton.  $V$  is the sample volume.  $g$  and  $f$  describe the exciton-photon and exciton-exciton interactions. In the many-exciton system the exciton-photon transition is assisted by the presence of an exciton other than that directly interacting with the photon. This exciton-assisted exciton-photon transition is described by terms proportional to  $l$  in (1) and represented by Feynman diagrams in figure 1, which can be interpreted as follows. A photon is absorbed generating an electron-hole pair, which does not form the final exciton state as in the usual exciton-photon transition. Instead, the electron (hole) of this pair is, due to the Pauli exclusion principle, exchanged with the electron (hole) of an exciton already existing in the system at the time of the photon absorption to form two final excitons as illustrated in figure 1 if it is read from left to right. Otherwise, if time is taken from right to left, one has the inverse process in which two excitons collectively generate a photon and an exciton. For three-dimensional samples the coupling constants  $g$ ,  $f$  and  $l$  can analytically be expressed as [23, 24] (for the two-dimensional case see, e.g., [28] and [29]).

$$g = -E\sqrt{\epsilon_0\Delta_{LT}/2w} \quad f = \frac{26}{3}\pi Ry r_x^3 \quad l = -7\pi g r_x^3 \quad (2)$$

with  $\epsilon_0$  the static dielectric constant of the semiconductor,  $\Delta_{LT}$  the exciton longitudinal-transverse splitting,  $Ry$  the exciton Rydberg and  $r_x$  the exciton Bohr radius. For bulk GaAs (CdS) one has  $E = 1.495$  (2.553) eV,  $\Delta_{LT} = 0.1$  (1) meV,  $Ry = 5$  (32.9) meV,  $\epsilon_0 = 12$  (8) and  $r_x = 100$  (25.5) Å, which give  $g \simeq -30$  (-101) meV,  $f \simeq 1.4 \times 10^{-16}$  ( $1.5 \times 10^{-17}$ ) meV cm<sup>3</sup> and  $l \simeq 6.6 \times 10^{-16}$  ( $3.7 \times 10^{-17}$ ) meV cm<sup>3</sup>. We see that  $f$  and  $l$  are of the same order of magnitude at least for bulk GaAs and CdS. In lower dimensions the expressions for  $f$  and  $l$  are very different from those given by (2) and they are difficult to calculate even numerically. Nevertheless, being of dipole-dipole-like character, the exciton-exciton interaction is weakened in low dimensions because each exciton becomes 'more neutral', i.e. less dipole like. On the other hand, the  $l$  interaction, by its nature, is less affected by 'exciton neutralization'. Thus, in low dimensions the  $l$  interaction is expected to be more important than the  $f$  one. In general, both interactions  $f$  and  $l$  give their own contributions, and they need to be considered simultaneously as they were in establishing the Hamiltonian (1).

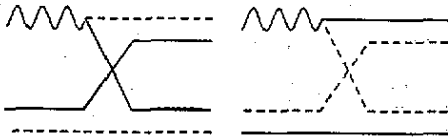


Figure 1. Feynman diagrams corresponding to the  $l$  interaction in the Hamiltonian (1). Full (broken) lines represent electron (hole) lines. A parallel pair of a full line and a broken one represents an exciton. An exchange of two electrons (holes) is described by a crossing of full (broken) lines. Photons are shown as wavy lines. No arrows are indicated: each diagram can be read from both sides. From left (right) to right (left) they correspond to photon absorption (emission).

### 3. Exciton quadrature variances

As in the problem of squeezed states of light, we define two exciton quadrature operators  $Q_\nu(t)$ , with  $\nu = 1$  or 2, as

$$Q_\nu(t) = \frac{1}{2}(i)^{\nu-1}[a^\dagger(t) - (-1)^\nu a(t)]. \quad (3)$$

Obviously,

$$[Q_\nu(t), Q_\mu(t)] = \frac{1}{2}i(-1)^\nu(\delta_{\nu\mu} - 1) \quad (4)$$

and

$$\langle [\Delta Q_1(t)]^2 \rangle \langle [\Delta Q_2(t)]^2 \rangle \geq \frac{1}{16} \quad (5)$$

where  $\Delta Q_\nu = Q_\nu - \langle Q_\nu \rangle$  and the average  $\langle \dots \rangle$  is understood in the quantum sense.  $\langle [\Delta Q_\nu(t)]^2 \rangle$  is referred to as exciton quadrature variances or briefly variances. Coherent states [3] are the ones in which both the variances are equal to  $\frac{1}{4}$ . These are states of minimal uncertainty because the variance product in (5) is exactly equal to  $\frac{1}{16}$ . Squeezed states are the minimal-uncertainty states in which, however, one of the two variances is less than  $\frac{1}{4}$ . If we use the normal-ordering symbol  $N$ , then in a squeezed state  $\langle N[\Delta Q_\nu(t)]^2 \rangle < 0$  for  $\nu = 1$  or 2. Since we shall assume the exciton-photon system to be in an initial coherent state whose amplitude is macroscopically large [3], it is convenient to deal with the normally ordered variances per unit volume, which are determined by

$$q_\nu(t) = (1/V) \langle N[\Delta Q_\nu(t)]^2 \rangle. \quad (6)$$

To analyse exciton squeezing we try to solve (1) for the time-varying exciton operators  $a(t)$  and  $a^+(t)$  and then examine the behaviour of  $q_\nu(t)$  to see whether or not they are negative during the course of time. For this purpose, we apply the Bogolubov transformations with real function coefficients [30, 31],

$$u_\nu = 1/[1 + g^2/(\Omega_\nu - E)^2]^{1/2} \quad v_\nu = gu_\nu/(\Omega_\nu - E) \quad (7)$$

where

$$\Omega_\nu = \frac{1}{2} \left[ w + E + (-1)^\nu \sqrt{(w - E)^2 + 4g^2} \right] \quad (8)$$

to bring (1) into a formula expressed in terms of the two new polariton operators  $\alpha_1$  ( $\alpha_1^+$ ) and  $\alpha_2$  ( $\alpha_2^+$ ):

$$H = \sum_\nu \Omega_\nu \alpha_\nu^+ \alpha_\nu + \frac{1}{V} \sum_{\nu\mu\xi\zeta} F_{\nu\mu\xi\zeta} \alpha_\nu^+ \alpha_\mu^+ \alpha_\xi \alpha_\zeta. \quad (9)$$

In (9) each of  $\nu$ ,  $\mu$ ,  $\xi$  and  $\zeta$  takes a value of either 1 or 2,

$$\alpha_\nu(t) = u_\nu c(t) + v_\nu a(t) \quad \alpha_\nu^+(t) = u_\nu c^+(t) + v_\nu a^+(t) \quad (10)$$

$$F_{\nu\mu\xi\zeta} = f u_\nu v_\mu v_\xi v_\zeta + l(v_\nu v_\mu v_\xi u_\zeta + u_\nu v_\mu v_\xi v_\zeta). \quad (11)$$

The transformations inverse to (10) read

$$a(t) = \sum_\nu u_\nu \alpha_\nu(t) \quad a^+(t) = \sum_\nu v_\nu \alpha_\nu^+(t) \quad (12)$$

$$c(t) = \sum_\nu u_\nu \alpha_\nu(t) \quad c^+(t) = \sum_\nu u_\nu \alpha_\nu^+(t). \quad (13)$$

To proceed further we now resort to the so-called secular approximation [32] which retains in the quadruple sum of (9) only the resonant terms proportional to  $F_{\nu\mu\nu}$ . The Hamiltonian can then be rewritten in the form

$$H = \sum_{\nu} \Omega_{\nu} \alpha_{\nu}^{\dagger} \alpha_{\nu} + \frac{1}{V} \sum_{\nu\mu} F_{\nu\mu} \alpha_{\nu}^{\dagger} \alpha_{\mu}^{\dagger} \alpha_{\mu} \alpha_{\nu} \tag{14}$$

where

$$\begin{aligned} F_{11} &= v_1^3 (f v_1 + 2lu_1) & F_{12} &= v_1 v_2^2 (f v_1 + 2lu_1) \\ F_{21} &= v_1^2 v_2 (f v_2 + 2lu_2) & F_{22} &= v_2^3 (f v_2 + 2lu_2). \end{aligned} \tag{15}$$

We note that (14) formally resembles a particular model Hamiltonian describing two photon coupled modes [33]. However, our Hamiltonian (14) is a concrete one with well specified parameters contained in  $F_{\nu\mu}$  through  $u_{\nu}$ ,  $v_{\nu}$  (see (7)) and  $f$ ,  $l$  (see (1)). This enables us in the next section to apply our theoretical results to a real semiconductor. The Heisenberg equation of motion set up from (14) for the operator  $n_{\nu}(t) = \alpha_{\nu}^{\dagger}(t)\alpha_{\nu}(t)$  is

$$d/dt n_{\nu}(t) = i[H, n_{\nu}(t)] = 0 \tag{16}$$

for both  $\nu = 1$  and  $2$ , i.e.  $n_{\nu}(t) = n_{\nu}(0) = \text{constant}$ . Then the Heisenberg equations of motion for  $\alpha_{\nu}^{\dagger}(t)$ ,

$$\frac{d}{dt} \alpha_{\nu}^{\dagger}(t) = i[\alpha_{\nu}^{\dagger}(t), H] = i\left(\Omega_{\nu} + \frac{1}{V} \sum_{\mu} (F_{\nu\mu} + F_{\mu\nu}) n_{\mu}(0)\right) \alpha_{\nu}^{\dagger}(t) \tag{17}$$

are easy to solve yielding explicitly analytical solutions

$$\begin{aligned} \alpha_{\nu}^{\dagger}(t) &= \alpha_{\nu}^{\dagger}(0) \exp \left[ i \left( \Omega_{\nu} + \frac{1}{V} \sum_{\mu} (F_{\nu\mu} + F_{\mu\nu}) n_{\mu}(0) \right) t \right] \\ \alpha_{\nu}(t) &= \exp \left[ -i \left( \Omega_{\nu} + \frac{1}{V} \sum_{\mu} (F_{\nu\mu} + F_{\mu\nu}) n_{\mu}(0) \right) t \right] \alpha_{\nu}(0). \end{aligned} \tag{18}$$

Substituting (18) into (12) gives the time dependence of the exciton operators, which we shall now use to evaluate the exciton quadrature variances  $q_{\nu}(t)$  in (6).

We anticipate that the exciton-photon system is initially in a coherent state denoted by

$$|x, Z_a; \gamma, Z_c\rangle = D_a(Z_a) D_c(Z_c) |x, 0; \gamma, 0\rangle \tag{19}$$

where  $D_b(Z_b)$  for any bosonic operator  $b$  is the displacement operator defined by

$$D_b(Z_b) = \exp(Z_b b^{\dagger} - Z_b^* b) \tag{20}$$

and  $Z_a, Z_c$  are arbitrary complex numbers characterizing the initial degree of coherence of the system. The average in (6) should be taken at time  $t > 0$ . In our case that means  $\langle \dots \rangle \equiv \langle \gamma, Z_c(t); x, Z_a(t) | \dots | x, Z_a(t); \gamma, Z_c(t) \rangle_t$ . Nevertheless, as seen from (12) and (18), the behaviours of all of the operators concerned are determined only by their behaviours at the initial time  $t = 0$ . We can thus write

$$\begin{aligned} \langle \dots \rangle &\equiv \langle \gamma, Z_c(t); x, Z_a(t) | \dots | x, Z_a(t); \gamma, Z_c(t) \rangle_t = \langle \gamma, Z_c; x, Z_a | \dots | x, Z_a; \gamma, Z_c \rangle_0 \\ &\equiv \langle \gamma, 0; x, 0 | D_c^{\dagger}(Z_c) D_a^{\dagger}(Z_a) \dots D_a(Z_a) D_c(Z_c) | x, 0; \gamma, 0 \rangle. \end{aligned} \tag{21}$$

Using (12) and (20) the following relations can be found:

$$D_a(Z_a) = \prod_{\nu} D_{\alpha_{\nu}}(v_{\nu}Z_a) \quad D_c(Z_c) = \prod_{\nu} D_{\alpha_{\nu}}(u_{\nu}Z_c) \quad (22)$$

which bring (21) into the form

$$\langle \dots \rangle = \langle \gamma, 0; x, 0 | \prod_{\nu} D_{\alpha_{\nu}}^+(u_{\nu}Z_c) D_{\alpha_{\nu}}^+(v_{\nu}Z_a) \dots \prod_{\nu} D_{\alpha_{\nu}}(v_{\nu}Z_a) D_{\alpha_{\nu}}(u_{\nu}Z_c) | x, 0; \gamma, 0 \rangle. \quad (23)$$

With this understanding of the average we are now in a position to analytically evaluate the exciton quadrature variances  $q_{\nu}(t)$ . First, using (6), (3), (12) and (18) we express the two variances  $q_{\nu}(t)$  in terms of the Bogolubov transformation functions and the polariton operators as

$$q_{\nu}(t) = \sum_{\xi\zeta} v_{\xi} v_{\zeta} X_{\xi\zeta}^{\nu}(t) \quad (24)$$

where

$$X_{\xi\zeta}^{\nu}(t) = (1/2V) \{ \text{Re}(\alpha_{\xi}^+(t)\alpha_{\zeta}(t)) + (-1)^{\nu-1} \text{Re}(\alpha_{\xi}(t)\alpha_{\zeta}(t)) \\ - 2 \text{Re}[(-i)^{\nu-1}(\alpha_{\xi}(t))] \text{Re}[(-i)^{\nu-1}(\alpha_{\zeta}(t))] \}. \quad (25)$$

Then, making use of the properties of the displacement operator we can prove the following equalities for arbitrary bosonic operator  $b$  and complex numbers  $Z_1, Z_2, \lambda$ :

$$\langle \gamma, 0; x, 0 | \dots b D_b(Z_1) D_b(Z_2) | x, 0; \gamma, 0 \rangle \\ = (Z_1 + Z_2) \langle x, 0; \gamma, 0 | \dots D_b(Z_1) D_b(Z_2) | x, 0; \gamma, 0 \rangle \quad (26)$$

and

$$\langle \gamma, 0; x, 0 | D_b^+(Z_2) D_b^+(Z_1) \exp(\lambda b^+ b) D_b(Z_1) D_b(Z_2) | x, 0; \gamma, 0 \rangle \\ = \exp\{[|Z_1|^2 + 2 \text{Re}(Z_1^* Z_2) + |Z_2|^2] \lambda\}. \quad (27)$$

At the last step, we put (18) into (25) and use (23), (26) and (27) to evaluate  $X_{\xi\zeta}^{\nu}(t)$ . With some trigonometric manipulations, we have obtained the following formulae (for simplicity  $Z_a$  and  $Z_c$  are assumed here to be real):

$$X_{\xi\zeta}^{\nu} = (-1)^{\nu} S_{\xi} S_{\zeta} \sin\{[P_{\xi\zeta} + (F_{\xi\zeta} + F_{\zeta\xi})/2V]t\} \sin\{[(F_{\xi\zeta} + F_{\zeta\xi})/2V]t\} \quad (28)$$

where the notations involved are

$$S_{\xi} = u_{\xi} \sqrt{z_c} + v_{\xi} \sqrt{z_a} \quad z_a = Z_a/V \quad z_c = Z_c/V \\ P_{\xi\zeta} = \Omega_{\xi} + \Omega_{\zeta} + \sum_{\mu} S_{\mu}^2 (F_{\mu\xi} + F_{\xi\mu} + F_{\mu\zeta} + F_{\zeta\mu}). \quad (29)$$

Substituting (28) into (24) gives the final, fully analytic expressions determining the exciton quadrature variances  $q_{\nu}(t)$ . Apart from the secular approximation, the derived expressions for  $q_{\nu}(t)$  are exact and look most compact (for comparison see [33] and [13]).

#### 4. Analysis of exciton squeezing over the parameter space

In this section the analytically derived formulae for the exciton quadrature variances, (24) with  $X_{\xi\xi}^v$  given by (28), will be graphically analysed over the relevant parameter space. There are two kinds of parameter: one is tunable and the other is dependent on a specific material whose properties will be determined experimentally. We call the former external and the latter internal. In reality, any physical effect depends simultaneously on a number of parameters, both external and internal. Furthermore, a change in one parameter might cause changes in other parameters, i.e. a parameter might be a function of many other parameters. All this makes an analysis over the parameter space not often an easy task. Here we shall carry out the analysis purely from a theoretician's point of view. That is, we assume that each parameter is an independent variable and only one or two parameters vary at a time while all the others are fixed. In the problem considered here, the external parameters are the photon frequency  $\omega$ , the initial degree  $Z_a, Z_c$  of coherence of the system and the size  $V$  of the experimental sample. Instead of the first two parameters we can equivalently consider the scaled frequency  $\omega/E$  and the degree  $z_a = Z_a/V, z_c = Z_c/V$  of coherence per unit volume. To the internal parameters belong  $E, g, f$  and  $l$ , which explicitly enter the model Hamiltonian (1). To be specific for the internal parameters let us choose GaAs as an example. Its parameters' values are provided in section 2.

In figure 2 we analyse the dependence of  $q_v = q_v(t)$  of GaAs on  $\omega/E$ , which is altered from 0.6 to 1. The other two external parameters are fixed as  $z_a = z_c = 1 \times 10^3 \text{ cm}^{-3}$  and  $V = 1 \times 10^{-15} \text{ cm}^3$ . Figure 2(a) for  $q_1$  and (b) (under a different angle of 3D view) for  $q_2$  displays negative peaks corresponding to the occurrence of exciton squeezing. The peaks are quite shallow (weak squeezing) for off-resonance frequencies  $\omega/E \leq 0.6$  and become deeper (stronger squeezing) for  $\omega/E$  approaching unity. The strongest squeezing is achieved for the perfect resonance detuning  $\omega = E$  (i.e.  $\omega/E = 1$ ) as clearly seen from figure 2. Figure 3(a) shows the time variation of  $q_v$  in dependence on  $n \equiv z_a = z_c = 0 \div 1000 \text{ cm}^{-3}$  and  $\omega/E = 1, V = 1 \times 10^{-15} \text{ cm}^3$ . The message from figure 3(a) is that the greater the initial coherence degree  $n$  the stronger the exciton squeezing. The peaks of  $q_1$  and  $q_2$  alternate in time exhibiting the fact that the two quadrature components can by no means be simultaneously reduced under the coherence-state values; otherwise, the Heisenberg uncertainty relations would be violated. The peaking alternation is most apparent in figure 3(b), where we plot 2D graphs for  $q_v = q_v(Et, V)$  with perfect resonance detuning and  $z_a = z_c = 1 \times 10^3 \text{ cm}^{-3}$ .  $q_1$  curves are dashed whereas  $q_2$  ones are solid. Curves with smaller oscillation amplitudes (weaker squeezing) corresponds to larger volumes  $V$  which are chosen to be equal to 1, 2, 5, 10 and  $50 \times 10^{-15} \text{ cm}^3$ . At  $t = 0$  both  $q_1$  and  $q_2$  are 'coherent'. Then  $q_1$  becomes 'squeezed' but  $q_2$  'expanded'. At some later moment of time they both return to coherent states; after that  $q_1$  turns out to be 'expanded' but  $q_2$  'squeezed', and so forth. At no moment of time are both  $q_1$  and  $q_2$  found as 'squeezed', thus defending the Heisenberg inequality.

To analyse exciton squeezing over the internal parameter space we reduce the number of parameters by one by considering the scaled quantities  $g/E, f/E$  and  $l/E$ . For convenience we reduce further the number of parameters by anticipating that  $f/E = l/E = L \times 10^{-21}$ . From the parameters' values given in section 2 for two typical semiconductors GaAs and CdS we can change  $|g|/E$  from 0 to 0.05 and  $L$  from 0 to  $100 \text{ cm}^3$ . To follow how exciton squeezing varies over  $|g|/E$  and  $L$  it is best to take moments of time corresponding to negative peaks. Two such moments, as can be seen from figure 3(b), are at  $Et = 13.3$  and  $Et = 15$ . Figure 4(a) shows  $q_1$  (lower 'squeezed' grid) and  $q_2$  (upper 'expanded' grid) in the space of  $L$  and  $E/g$  at  $Et = 13.3$  and for  $\omega/E = 0.9, z_a = z_c = 1 \times 10^3 \text{ cm}^{-3}$  and



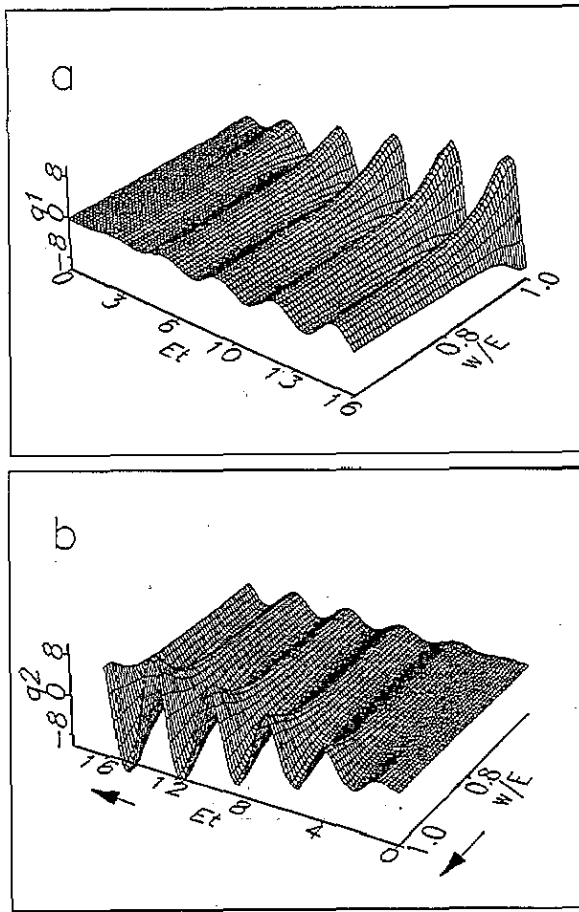


Figure 2. GaAs time-varying exciton quadrature variances  $q1 \equiv q_1$  (a) and  $q2 \equiv q_2$  (b) versus scaled photon frequency  $w/E$ .  $n_a = n_c = 1 \times 10^{15} \text{ cm}^{-3}$ ,  $V = 1 \times 10^{-15} \text{ cm}^3$ . Negative peaks signal the occurrence of exciton squeezing.

$V = 1 \times 10^{-15} \text{ cm}^3$ . Figure 4(b) is the same as figure 4(a) except that  $Et = 15$ : here however the lower 'squeezed' (upper 'expanded') grid represents  $q_2(q_1)$ ! From figure 4(a) and (b) we learn that if either  $g/E = 0$  or  $L = 0$  no exciton squeezing can occur: the two grids coincide at the line  $q_{1,2} = 0$ ,  $g/E = 0$  and the line  $q_{1,2} = 0$ ,  $L = 0$ . This result is physically understandable: being optically excited and non-linear in nature, squeezed excitons could not be generated if the light-matter interaction is turned off ( $g = 0$ ) and/or no non-linearities are included ( $L = 0$ ). Therefore, the necessary conditions for excitons to be squeezed are simultaneous  $g \neq 0$  and  $L \neq 0$  (i.e.  $f$  and/or  $l \neq 0$ ). It is interesting to note here that this can immediately be gathered in an analytic way from (28). Indeed, if  $g = 0$  ( $L = f = l = 0$ ) then  $v_v = 0$  as follows from (7) (the parentheses in (15) are equal to zeros). Once  $v_v = 0$  (the parentheses in (15) are equal to zeros) all the  $F_{\xi\xi}$  in (15) also vanish making the second sinusoidal function in (28) zero, too. This yields  $X_{\xi\xi}^v(t) = 0$ , i.e.  $q_v = 0$  all the time, which in turn means the absence of exciton squeezing. Figure 4(a) and (b) signals a better exciton squeezing for materials with larger interactions. Summing up the analysis results we would conclude that it is possible to produce squeezed excitons in a small-volume semiconductor with large interactions ( $g$ ,  $f$  and/or  $l$ ) pumped by a coherent light of frequency close to the exciton level.

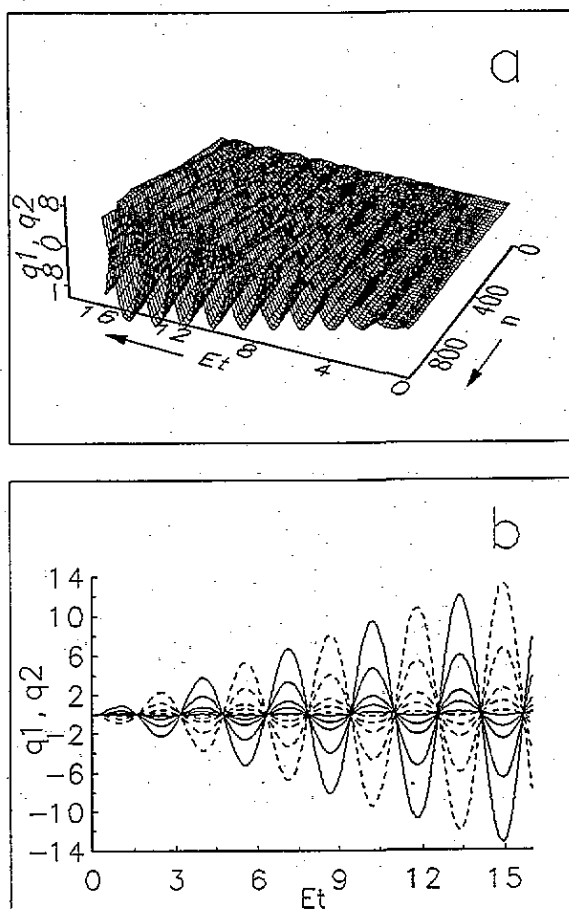


Figure 3. As figure 2 but (a) versus  $n \equiv z_a = z_c$  in  $\text{cm}^{-3}$ ;  $w/E = 1$  and  $V = 1 \times 10^{-15} \text{ cm}^3$ ; (b) versus  $V$ ;  $w/E = 1$  and  $z_a = z_c = 1 \times 10^3 \text{ cm}^{-3}$ .  $q_1$  curves are broken,  $q_2$  ones are full. Smaller-oscillation amplitudes correspond to  $V = 1, 2, 5, 10$  and  $50 \times 10^{-15} \text{ cm}^3$ , respectively.

## 5. Discussion

We now discuss three points. The first concerns the validity of the model Hamiltonian used (1). This Hamiltonian can be rigorously derived from the original photon-electron-hole picture by mapping onto a hypothetical bosonic space with the aid, e.g., of Usui's transformations [34] (see [23] and [24]). This and the expressions (2) are correct as far as the exciton density  $\rho_x$  satisfies the condition [23, 24]  $26\pi r_x^3 \rho_x / 3 \ll 1$ . Taking  $r_x = .50 \text{ \AA}$  as in typical semiconductors,  $\rho_x$  must be much less than  $2.9 \times 10^{17} \text{ cm}^{-3}$ . In our coherent system the quantity equivalent to exciton density is  $z_a$ . Since in our numerical calculations we took  $z_a$  as small as  $z_a = 0 \div 10^3 \text{ cm}^{-3}$ , the validity of the working Hamiltonian (1) is surely justified.

The second point to be discussed regards the size dependence. From figure 3(b) it follows that the effect of exciton squeezing should not manifest itself in large-volume samples. We analysed  $V$  of about some or some tens of  $10^{-15} \text{ cm}^3$ . Suppose the sample is a sphere of radius  $R$ . The analysed values of  $V$  give  $R \simeq 10^3 \text{ \AA} \simeq 20r_x$ . These are intermediate between bulk samples and microcrystallites [35]. The dependence of statistical

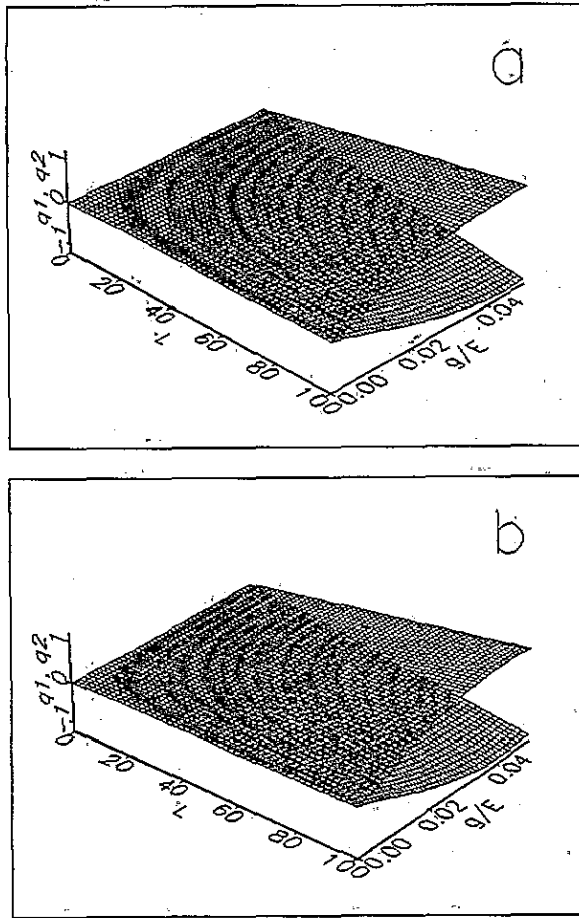


Figure 4. Exciton quadrature variances  $q_v$  in the parameter space of  $L$  (for  $L$  see text) and  $|g|/E$  (note that  $g/E$  in the figure should be read as  $|g|/E$ ).  $w/E = 0.9$  and  $V = 1 \times 10^{-15} \text{ cm}^3$ . (a)  $Et = 13.3$ . The upper (lower) grid is for  $q_2$  ( $q_1$ ). (b)  $Et = 15$ . The upper (lower) grid is for  $q_1$  ( $q_2$ ).

properties on the volume in crystals of such intermediate sizes is reported in [10] and [11], whereas several physical phenomena were predicted to be enhanced in microcrystallites due to the finite-size effect [36]. Therefore, the above-reported exciton squeezing effect is also hoped to be enhanced in small quantum samples. To confirm this, however, needs further investigations because in such quantum microcrystallites  $E$ ,  $g$ ,  $f$  and  $l$  in (1) require highly complicated evaluations, which are difficult even numerically (for instance see an effective Hamiltonian for large microcrystallites in [37]).

Coming to the end we wish to say some words about how to 'detect' squeezed excitons. Discussions on experimentally detecting the squeezed polariton structure were given in [9]. Yet, polaritons as well as excitons are intracrystal elementary excitations, and no devices can be put inside a sample to directly detect them in a squeezed state. One may only indirectly check them outside the experimental sample through specific effects that are consequences of their squeezing. As shown in [12], in a light-matter coupled system excitons and photons are able to transfer their statistical properties from one to another. Squeezed behaviour of excitons should therefore be transferred to intracrystal photons and vice versa. Since photons

propagate through and leave the sample, they can be detected outside the sample. Hence, an observation of squeezed photons behind a semiconductor sample could serve as possible evidence of the presence of squeezed excitons inside the semiconductor. In fact, in the spectral region near the semiconductor band edge, exciton squeezing and photon squeezing are two closely connected problems. In a similar fashion, photon squeezing via an excitonic non-linearity has recently been treated in [38]. In separate research we will establish and clarify the relationship between squeezed states of excitons, photons and polaritons inside a semiconductor.

Finally, we wish to recall that the results obtained in this paper were based on a very strong idealization of real systems in writing the effective Hamiltonian (1). More realistic models are worth investigating but involve rather complicated theoretical evaluations, which we hope will not change the qualitative picture:

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