

Exciton squeezed state in optically excited semiconductors

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Current pump-probe techniques allow one to prepare highly excited semiconductors which can be modelled as interacting exciton–photon systems. We theoretically evaluate the exciton quadrature variances for such systems which show that the initially coherent exciton can periodically evolve into a squeezed state. Necessary constraints for the squeezing effect are found and an analysis over the parameter space is given.

Following successful investigations of nonclassical effects of light in quantum optics [1], interest in such effects has recently arisen also in quantum field theory [2,3] and in condensed matter physics [4–11]. Soliton squeezing was studied in refs. [2–4]. Reference 5 confirmed that squeezed phonons give a lower ground-state energy of the whole superconducting system than coherent and displaced ones do. An isomorphism between a polariton model Hamiltonian and a standard two-mode squeeze Hamiltonian was established in ref. [6] where the wavevector dependence of the squeezing factor of the polariton was given. References [7] and [8] dealt with the statistical properties of the coherently excited polariton wave in small volume crystals. Squeezed states of excitons and biexcitons were considered respectively in refs. [9], [10] and [11].

Under a resonant optical pulse excitation a finite population of coherent excitons can be generated in a semiconductor. These excitons are then optically tested by a weak probe beam of photons with frequency lying in the vicinity of the exciton energy

level. If the excitation is low, the optical properties of the exciton–photon system are governed by the Hamiltonian ($\hbar=c=1$)

$$H = \omega c^+ c + E a^+ a + g(a^+ c + c^+ a), \quad (1)$$

where E (ω) is the energy (frequency) of the coherent exciton (photon) whose operators are denoted by a , a^+ (c , c^+), g is the exciton–photon coupling constant. In this case, as shown in ref. [9], the exciton can evolve into a squeezed state only if the photon initially possesses a certain degree of squeezing. At higher excitation, a finite exciton population is created which may induce qualitative modifications of the optical properties. There arise nonlinear exciton–photon transitions (see fig. 1) due to exchange polarization [12,13], which are described by terms proportional to $a^+ a^+ a c + c^+ a^+ a a$. Besides, as a rule, the exciton–exciton interaction comes into play, too. The full Hamiltonian therefore has the form

$$H = \omega c^+ c + E a^+ a + g(a^+ c + c^+ a) + \frac{f}{V} a^+ a^+ a a + \frac{l}{V} (a^+ a^+ a c + c^+ a^+ a a), \quad (2)$$

where V is the sample volume. A particular case of

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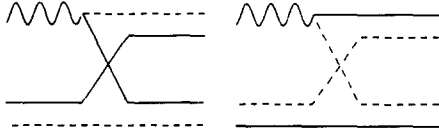


Fig. 1 Feynman diagrams corresponding to the l -interaction in Hamiltonian (2). Solid (dashed) lines represent electron (hole) lines. A parallel pair of a solid line and a dashed one represents an exciton. An exchange of two electrons (holes) is described by a crossing of solid (dashed) lines. Photons are shown as wiggly 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).

(2) with $l=0$ was treated in ref. [10] where the f -term was shown to cause the appearance of squeezed excitons even when the system is initially in a non-squeezed state. In this Letter we will prove that f - and l -terms play equal roles in the squeezing phenomenon: squeezing might occur when either f or l is non-zero. In general, for bulk semiconductors, f and l are of the same order of magnitude and, thus, they need to be taken into account simultaneously.

To study exciton squeezing we will evaluate the time-dependence of the exciton normally-ordered quadrature variances $Q_\nu(t)$ defined for $\nu=1, 2$ as

$$Q_\nu(t) = \frac{1}{4} (-1)^{\nu-1} \langle N \{ \Delta [a^+(t) - (-1)^\nu a(t)] \}^2 \rangle, \quad (3)$$

where the symbol N normally orders all the operators standing after it, $\Delta x = x - \langle x \rangle$ and $\langle \rangle$ is the quantum average. One says the exciton is squeezed when either $Q_1(t)$ or $Q_2(t)$ becomes negative. Applying the Bogolubov transformations which diagonalize the quadratic part of (2) into the two-branch polariton [14,15] with energies Ω_1 and Ω_2 and, making use of the secular approximation [16], we have obtained the following analytically exact expressions for $Q_\nu(t)$,

$$Q_\nu(t) = (-1)^\nu \sum_{\xi, \zeta=1}^2 v_\xi v_\zeta S_\xi S_\zeta \times \sin \left[\left(P_{\xi\zeta} + \frac{F_{\xi\zeta} + F_{\zeta\xi}}{2V} \right) t \right] \sin \left(\frac{F_{\xi\zeta} + F_{\zeta\xi}}{2V} t \right). \quad (4)$$

In (4) the notations used are

$$S_\xi = u_\xi \sqrt{z_c} + v_\xi \sqrt{z_a}, \quad z_a = Z_a/V, \quad z_c = Z_c/V, \quad (5)$$

$$P_{\xi\zeta} = \Omega_\xi + \Omega_\zeta + \sum_{\mu} S_\mu^2 (F_{\mu\xi} + F_{\xi\mu} + F_{\mu\zeta} + F_{\zeta\mu}), \quad (6)$$

$$F_{\xi\zeta} = v_\xi v_\zeta (f v_\xi + 2l u_\xi), \quad (7)$$

where u_ξ and v_ξ are the Bogolubov transformation coefficients, Z_a and Z_c , which are for simplicity assumed real in the evaluation of (4), characterize the initial coherence degree of the exciton and photon, respectively. While the detailed derivation of (4) will be published elsewhere, we wish here to stress its three most qualitatively impressive consequences.

(i) Formula (4), though simple in form, contains an explicit dependence on both external (ω , V , Z_a , Z_c) and internal (E , g , f , l) parameters (note that ω and g enter u_ξ , v_ξ and Ω_ξ whose analytic expressions are given e.g. in ref. [15]). This allows us to analyze easily the variation of exciton squeezing over the parameter space in figs. 2 and 3. (ii) The presence of the second sinus in (4) reveals that $Q_\nu(t)$ is identically equal to zero, i.e. the exciton remains in its initial coherent state and no squeezing can take place, if $F_{\xi\zeta}=0$ for all possible ξ and ζ . Looking at (7) we find out that this will occur when either $f=l=0$ or $v_\xi=0$ for both $\xi=1$ and 2. Since $v_\xi \propto g$ (see their expressions in ref. [15]), we can theoretically conclude that the necessary constraints for the exciton to be squeezed are simultaneous nonvanishing of both g and l or f (not necessarily both l and f are different from zero). In low dimensions f turns out to be very small whereas l remains finite. In such circumstances l plays the key role in the squeezing problem. The inclusion of l into consideration from the very onset as we have done is thus meaningful. (iii) The prefactor $(-1)^\nu$ in (4) indicates that when

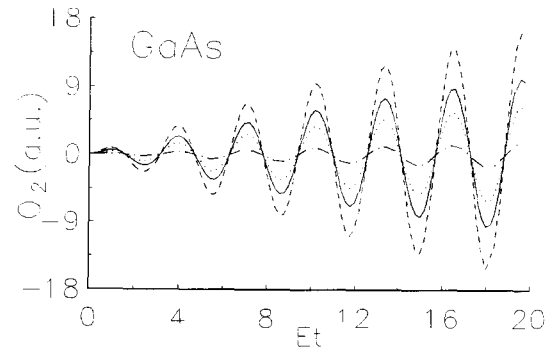


Fig. 2. GaAs time-dependent exciton quadrature variance Q_2 as influenced by the external parameters. For the values of the external parameters used see text.

Q_1 (Q_2) is negative, Q_2 (Q_1) should be positive. This guarantees the validity of the Heisenberg uncertainty relation: in no cases both Q_1 and Q_2 are negative.

Figure 2 plots Q_2 for GaAs as a function of Et with different external parameters. From the analytic formulas for g, f, l (see e.g. ref. [17]) and from concrete values of the internal parameters for GaAs we have $|g| \approx 30$ meV, $f \approx 1.4 \times 10^{-16}$ meV cm³ and $l \approx 6.6 \times 10^{-16}$ meV cm³. The dashed (solid, dotted, dot-dashed) curve is for $\omega/E=1$ (0.95, 1, 1), $z_a=z_c=1000$ (1000, 400, 1000) cm⁻³ and $V=10^{-15}$ (10^{-15} , 10^{-15} , 10^{-14}) cm³. Visually, the curves during the course of time develop negative parts indicating the appearance of squeezed excitons. Comparing the dashed and the solid (the dashed and the dotted, the dashed and the dot-dashed) curves indicates that a better squeezing is obtainable for ω closer to E (larger initial coherence degree, smaller sample volume). The chosen sample volumes of 10^{-15} and 10^{-14} cm³ are somewhat intermediate between bulk samples and microcrystallites [18]. The finite sample volume gives rise to the squeezing effect. This is in agreement with the result discussed in ref. [8] on the influence of the quantum fluctuation on the coherent system: in the limit $V \rightarrow \infty$ the influence is negligible.

To follow the exciton squeezing variation over the internal parameter space let us fix $\omega/E=0.9$, $z_a=z_c=1000$ cm⁻³, $V=10^{-15}$ cm³ and change E, g, f, l . Analyzing the parameters of typical semiconductors, we can vary $|g|/E$ from 0 to 0.05 and $X=(l/E) \times 10^{21}$ cm³ $\approx (f/E) \times 10^{21}$ cm³ from 0 to 100. In fig. 3 we draw Q_2 versus X and $|g|/E$ at $Et=15$ which corresponds to the fifth negative peak in fig. 2. As theoretically stressed above in (ii) and now seen from fig. 3, no squeezing arises when either g or X vanishes. Stronger linear ($\propto g$) and nonlinear ($\propto X$) interactions more favor the squeezing phenomenon.

Since excitons and photons are coupled to each other (by means of g and l), their statistical properties may be exchanged during the time-evolution. Therefore, the photon in the interacting exciton-photon system can also evolve into a squeezed state. A new mechanism of photon squeezing via exciton-exciton interaction was for the first time suggested in ref. [19].

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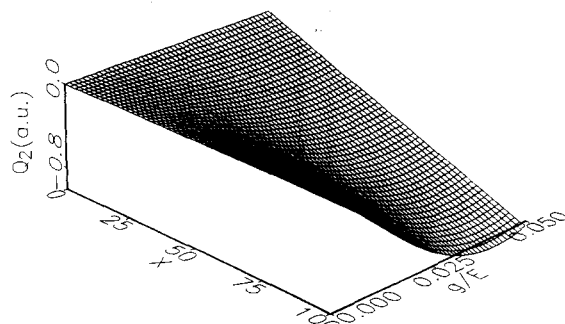


Fig. 3. Exciton quadrature variance Q_2 as influenced by the internal parameters. For the values of the external parameters used and for the definition of X see text. The label g/E in this figure should be understood as $|g|/E$.

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