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## DENSITY-DEPENDENT SQUEEZING OF EXCITONS IN HIGHLY EXCITED SEMICONDUCTORS

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### ABSTRACT

The time evolution from coherent states to squeezed states of high density excitons is studied theoretically based on the boson formalism and within the Random Phase Approximation. Both the mutual interaction between excitons and the anharmonic exciton-photon interaction due to phase-space filling of excitons are taken into account. It is shown that the exciton squeezing depends strongly on the exciton density in semiconductors and becomes smaller with increasing the latter.

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Recently the quantum optical properties of the quasiparticles in condensed matters have been investigated intensively [1-10]. In particular, the nonclassical properties of exciton-polariton were considered in [1-3], phonon-polariton in [4], and squeezed states of excitons were considered in [5-7]. Of particular interest are also the optical nonlinearities emerged from high external excitation which affect almost all the optical properties of semiconductors. At high excitation a great number of excitons can be produced and there arise some kinds of anharmonicities such as nonlinear exciton-exciton interaction and the anharmonic exciton-photon interaction due to phase-space filling of excitons. An attempt to consider the effect of these anharmonicities on squeezed states of excitons was first made in [5-7]. However, in [5,6] to solve the problem the authors had to resort to rough approximation neglecting many terms in Hamiltonian which actually should be retained. It is well known that for the high density systems where the fluctuations of the occupation number can be neglected, the Random Phase Approximation (RPA) [11] is the most suitable, and one can safely apply it to study nonlinear optical properties of the system. In this paper, as in [7], based on the boson formalism and within the framework of RPA, we study the effect of both above-mentioned kinds of anharmonicities on the squeezing properties of high density excitons. It will be shown that the squeezing degree of excitons depends strongly on the exciton density in semiconductors, and moreover, it decreases with increasing the latter.

We shall start with the following effective boson Hamiltonian describing the exciton-photon system at high excitation [9]

$$H_{eff} = E^{ex} a^+ a + \omega^\gamma c^+ c + f a^+ a^+ a a + [g_o c a^+ (1 - \lambda a^+ a) + \text{h.c.}], \quad (1)$$

where  $a^+(a)$ ,  $c^+(c)$  are the creation (annihilation) operators for exciton and photon;  $E^{ex}$ , and  $\omega^\gamma$  are the energies of the exciton and photon, respectively,  $g_o$  is photon-exciton coupling constant. The third term on the right-hand side of (1) describes the repulsive exciton-exciton interaction, and  $f$  is given by

$$f = -\frac{2}{V^3} \sum_{k, k'} V(k - k') [|\varphi(k)|^2 |\varphi(k')|^2 - |\varphi(k)|^2 \varphi(k) \varphi(k')]. \quad (2)$$

The fourth term on the right-hand side of (1) represents the anharmonic exciton-photon interaction due to phase-space filling of excitons, and  $\lambda$  is given by

$$\lambda = \frac{1}{V} \sum_k |\varphi(k)|^2 \varphi(k) / \sum_k \varphi(k). \quad (3)$$

Here  $V$  is the quantization volume,  $\varphi(k)$  - is the Fourier transform of the exciton envelope function for electron-hole relative motion,  $V(k - k')$  - is Coulomb potential.

We shall not detail here the derivation of Hamiltonian (1) we remind only that it is obtained by operating Usui transformation [12] on the Hamiltonian of the interacting photon-electron-hole system. This procedure was originally developed by the authors of [13,14] for boson formalism of high density exciton theory.

Let us consider the following Green's functions

$$G_1(k, t) = \theta(t) \langle [a(t), a^\dagger(o)] \rangle \equiv \langle \langle a(t), a^\dagger(o) \rangle \rangle, \quad (4)$$

$$G_2(k, t) \equiv \langle \langle c(t), a^\dagger(o) \rangle \rangle. \quad (5)$$

The equations of motion for the Green's functions (4,5) can be derived in standard way. Then using the RPA-type factorization which is reasonable for high densities, we obtain the closed system of equations, the solutions of which give us the spectrum of the exciton-polariton, as follows

$$\epsilon_{1,2} = \frac{1}{2}(E^* + \omega^\gamma) \pm \frac{1}{2}\sqrt{(E^* - \omega^\gamma)^2 + 4g^{*2}}, \quad (6)$$

where

$$E^* = E^{ex} + 2n_{ex}Vf, \quad (7)$$

$$g^{*2} = g_o^2(1 - n_{ex}V\lambda), \quad (8)$$

and  $n_{ex}$  is the density of excitons created by laser field in semiconductors.

The polariton whose spectrum is (6) can be now expressed in terms of photon  $c$  and "real" exciton  $\tilde{a}$  operators, as follows

$$B_i = u_i c + v_i \tilde{a}, \quad (9)$$

where

$$u_i = [1 + g^{*2}(\epsilon_i - E^*)^{-2}]^{-1/2}, \quad (10)$$

$$v_i = g^*|\epsilon_i - E^*|^{-1}u_i. \quad (11)$$

To analyse the time evolution of the squeezing properties of the exciton field we introduce two Hermitian quadrature exciton operators

$$X(t) = \frac{1}{2}[\tilde{a}^+(t) + \tilde{a}(t)], \quad (12)$$

$$P(t) = \frac{i}{2}[\tilde{a}^+(t) - \tilde{a}(t)]. \quad (13)$$

From (9) and the exact time-dependent expression for operator  $B_i(t)$  one can obtain the following equation

$$\tilde{a}(t) = \sum_{i=1,2} [u_i v_i c(o) + v_i^2 \tilde{a}(o)] e^{-i\epsilon_i t}. \quad (14)$$

Since the squeezed states of exciton are defined (analogous to light) as the states with a variance in one quadrature exciton operator smaller than that associated with the coherent state, the exciton becomes squeezed if

$$\langle : (\Delta X(t))^2 : \rangle = \langle (\Delta X(t))^2 \rangle - \frac{1}{4} < 0, \quad (15)$$

$$\text{or} \quad \langle : (\Delta P(t))^2 : \rangle = \langle (\Delta P(t))^2 \rangle - \frac{1}{4} < 0, \quad (16)$$

where  $\langle : (\Delta X(t))^2 : \rangle$  and  $\langle : (\Delta P(t))^2 : \rangle$  are so-called the normally ordered variances and are widely accepted as a measure of squeezing [15,16].

Let us, for simplicity, suppose that at initial time  $t = o$  excitons are in coherent state and light is prepared in a squeezed state characterized by the squeezed factor  $r$  and a phase  $\varphi$  [1]. From (9), (12)-(14) and after some algebraic manipulations we obtain the following exact analytical expressions for the exciton normally ordered variances

$$\langle : (\Delta X(t))^2 : \rangle = \frac{1}{2}|\beta'(t)|^2 \sinh^2 r - \frac{1}{2}\text{Re}(\beta'^2(t)e^{i\varphi}) \cosh r \sinh r, \quad (17)$$

$$\langle : (\Delta P(t))^2 : \rangle = \frac{1}{2}|\beta'(t)|^2 \sinh^2 r + \frac{1}{2}\text{Re}(\beta'^2(t)e^{i\varphi}) \cosh r \sinh r, \quad (18)$$

where

$$|\beta'(t)|^2 = 4u_1^2 v_1^2 \sin^2 \frac{(\epsilon_1 - \epsilon_2)t}{2}, \quad (19)$$

$$\text{Re}(\beta'^2(t)e^{i\varphi}) = -4u_1^2 v_1^2 \sin^2 \frac{(\epsilon_1 - \epsilon_2)t}{2} \cos(\epsilon_1 + \epsilon_2 - \varphi)t. \quad (20)$$

where  $\epsilon_1, \epsilon_2$  and  $u_i, v_i$  are defined above by (6)-(8) and (10),(11), respectively.

The formulas (17)-(20) are our main results from which we would like to emphasize the following. Since  $u_i, v_i$  are expressed in  $\epsilon_i, E^*$  and  $g^*$  (see (10),(11)) which in turn depend explicitly on light frequency  $\omega$ , coupling constants  $f, \lambda$ , and the exciton density  $n_{ex}$  (see (6)-(8)), one can expect various kinds of dependence of the exciton squeezing on these parameters.

Note that the exciton density  $n_{ex}$  always comes along with the parameters  $f$  and  $\gamma$  characterizing the nonlinear exciton-exciton interaction and anharmonicity of exciton-photon coupling due to phase space filling of exciton. The coupling constants  $f$  and  $\gamma$

were calculated for the lowest 1S-exciton in bulk semiconductors in [9,14]

$$f = \frac{13\pi}{3V} E_B a_B^3, \quad \gamma = \frac{7\pi}{2V} a_B^3. \quad (21)$$

where  $a_B$  is the effective Bohr radius of the bulk exciton and  $E_B$  is its binding energy.

For graphical illustration we take the case of CdS with the parameters  $E^{ex} = 2553\text{meV}$ ,  $g_o = 50\text{meV}$ ,  $a_B = 2.8 \times 10^{-7}\text{cm}$ ,  $E_B = 27.8\text{meV}$  and  $\varphi = 0, r = 0.8$ , scaled time is equal to  $E^{ex}t$ . To see the frequency dependence of the exciton squeezing, in Fig. 1a, b, c we plot the time evolution of the exciton normally ordered variance  $\langle (\Delta x(t))^2 \rangle$  for the fixed exciton density  $n_{ex} = 1.10^{17} \text{ cm}^{-3}$ , and three different values of photon energy  $\Delta\omega = E^{ex} - \omega = 350\text{meV}$ ,  $100 \text{ meV}$  and  $0 \text{ meV}$ , respectively. One can see that with photon frequency approaching closer to the exciton resonance ( $\Delta\omega \rightarrow 0$ ) the squeezing degree obtained by excitons from photons effectively increases, and gains its maximum at exact exciton resonance (Fig. 1c).

One of the new main results in this paper is explicit dependence of exciton squeezing on the exciton density. This is a consequence of the effects of nonlinear interaction between excitons and the anharmonic exciton-photon interaction due to phase space filling of excitons. Higher exciton densities screen more the interaction between excitons and photons, and reduce it by the factor  $(1 - n_{ex}V\lambda)$ . This is just the saturation effect due to phase space filling of excitons which usually appears in highly excited systems. The above-analysed density dependence of exciton squeezing is well seen from Fig. 2a, b, c plotted for the fixed photon frequency near exciton resonance  $\Delta\omega = 3 \text{ meV}$ , and some different exciton densities  $n_{ex}$ .

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## Figure Captions

**Fig.1** Time evolution of the exciton normally ordered variance  $\langle : (\Delta x(t))^2 : \rangle$  for  $n_{ex} = 1.10^{17} \text{ cm}^{-3}$ ;  $\Delta\omega = 350 \text{ meV}$  (a),  $100 \text{ meV}$  (b) and  $0 \text{ meV}$  (c). The closer to exciton resonance the greater exciton squeezing achieved.

**Fig.2** Time evolution of the exciton normally ordered variance  $\langle : (\Delta x(t))^2 : \rangle$  for  $\Delta\omega = 3 \text{ meV}$ ;  $n_{ex} = 1.10^{17} \text{ cm}^{-3}$  (a),  $1.10^{18} \text{ cm}^{-3}$  (b) and  $3.10^{18} \text{ cm}^{-3}$  (c). With increasing  $n_{ex}$  the exciton squeezing degree decreases.

**Fig.1**

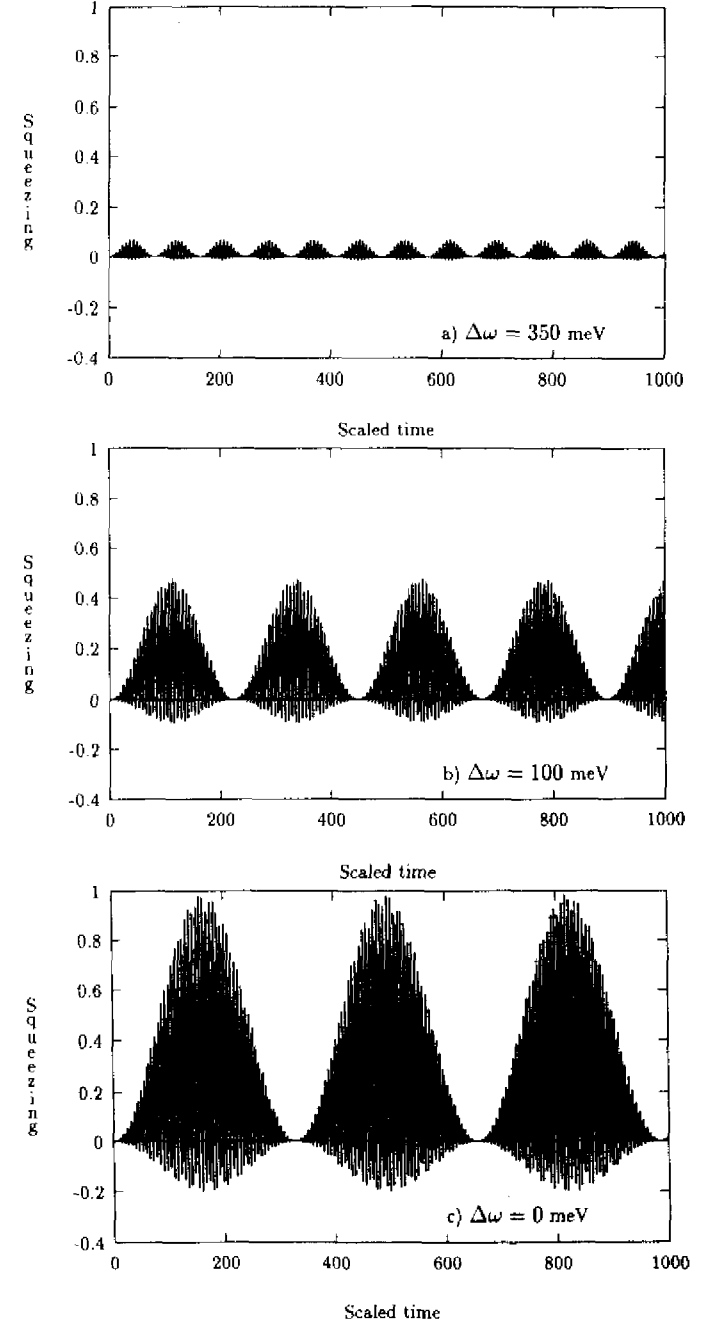


Fig.2

