

NONBOSON TREATMENT OF EXCITONIC NONLINEARITY IN OPTICALLY EXCITED MEDIA

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The present article shortly reviews some recent results in the study of excitonic nonlinearity in optically excited media using a nonboson treatment for many-exciton systems. After a brief discussion of the exciton nonbosonity the closed commutation relations are given for exciton operators which hold for any exciton density and type. The nonboson treatment is then applied to the problems of intrinsic optical bistability and nonlinear polariton yielding quite interesting and new effects, e.g. new shapes of hysteresis loops of intrinsic optical bistability or anomalies of polariton dispersion.

1. Introduction

It is not exceptional anymore that high density systems of composite quasiparticles like Cooper-pairs, excitons or biexcitons obey neither Bose- nor Fermi statistics. This feature brings about great difficulty in studying different physical phenomena with such quasiparticles involved. A way to overcome the difficulty is to search for a representation in which operators of the quasiparticles could be expanded into an infinite series in bosonic or fermionic operators.¹⁻⁴ Another way to describe many-exciton systems is to transform the original Hamiltonian in the real space where only true bosons and fermions exist in an effective Hamiltonian in a hypothetical space where all the fermions are bound into the composite quasiparticles which now behave as “hypothetical ideal bosons”.⁵⁻¹⁴ The latter way is in a sense also an expansion of a quasiparticle operator into an infinite or finite series in hypothetical bosonic operators (see, e.g., Ref. 10). In practical calculations we cannot handle long series and always have to limit ourselves just to few first terms of the series expansion. That means that results obtained within the above said restriction would be reliable only for low density cases.⁶ For the low density limit the electron-hole pairing procedure performed in Ref. 11 seems quite simple but rigorous to cast the original fermionic Hamiltonian into the effective Hamiltonian which is expressed only in terms of bosonic operators. In Ref. 14 the pairing procedure has been further applied to take into account the particle spin effect and thus the explicit spin-dependent interaction Hamiltonian in terms of exciton operators and the

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analytical investigation of the exciton-exciton interaction potential in both 2D and 3D materials have been derived and done therein for the first time.

To develop a formalism valid for any density it can be thought that composite quasiparticle operators must not be expanded into a series in bosonic operators, i.e. a nonboson approach is needed. For this purpose one, however, has to look for a closed set of commutation relations of the quasiparticle operators which might not be of the form of usual commutators or anticommutators as for true bosons or fermions and should hold validly for arbitrary density. Such a nonboson formalism was indeed developed in Refs. 15 and 16 where closed specific (bilinear and trilinear) commutation relations were found for operators of Frenkel-type excitons in molecular crystals. For Wannier-type excitons in semiconductors things seem much more confusing because instead of simple exponent functions^{15,16} we have to handle very complicated (e.g. hydrogen-like) ones.⁵⁻¹⁴ In Ref. 17 we have extended the results of Refs. 15 and 16 to the case of Wannier-type excitons. And, how surprising it may be, we have shown that our results are generalized and they can be used for any kind of excitons in materials of bulk or low-dimensional structure as well as under the influence of an external action. In this article we briefly remind the main results of Ref. 17 (Sec. 2) and then apply them to two important questions of the physics of highly excited materials. One of them, the optical bistability phenomenon (Sec. 3), is of potential applicability to future ultrafast digital computing. And, the other, nonlinear polariton dispersion (Sec. 4), plays a significant role in both explaining and predicting many optical processes, because polaritons are real modes that exist in optically excited media and their dispersion laws can be exactly derived by solving concrete light-matter interaction Hamiltonians.

Throughout the article we for convenience shall use the units of system with \hbar (Planck constant) = c (light velocity) = V (sample volume) = 1.

2. Closed Commutation Relations of Exciton Operators

As a kind of composite quasiparticles, an exciton is a bound state of an electron and a hole. Then the creation operator of excitons $b_{\mathbf{v}\mathbf{k}}^+$ can be constructed through those of free electrons $e_{\mathbf{k}-\mathbf{p}}^+$ and free holes $h_{\mathbf{p}}^+$:

$$b_{\mathbf{v}\mathbf{k}}^+ = \sum_{\mathbf{p}} f_{\mathbf{v}}(\mathbf{p} - \sigma\mathbf{k}) e_{\mathbf{k}-\mathbf{p}}^+ h_{\mathbf{p}}^+ \quad (1)$$

where \mathbf{k} and \mathbf{p} are quasimomenta, functions $f_{\mathbf{v}}$ characterize the relative electron-hole motion in an exciton and σ is the relative hole-exciton effective mass ratio. At low density $f_{\mathbf{v}}$ are simple exponential (or hydrogen-like) functions for Frenkel-type^{15,16} (or Wannier-type⁵⁻¹⁷) excitons. But at high density these functions must be selfconsistently determined by, e.g., a variational method¹¹ because they themselves depend on density.

It is easy to verify, on the one hand, that operators defined by Eq. (1) are not true boson nor fermion operators, i.e.

$$[b_1, b_2^+] \equiv b_1 b_2^+ - b_2^+ b_1 \neq \delta_{1,2} \quad (2)$$

$$\{b_1, b_2^+\} \equiv b_1 b_2^+ + b_2^+ b_1 \neq \delta_{1,2} . \quad (3)$$

In Eqs. (2) and (3) abbreviations 1 and 2 stand for $v_1 k_1$ and $v_2 k_2$.

On the other hand, one can try some algebra and finds the following closed set of quite specific commutation relations for exciton operators, namely

$$[[b_1, b_2^+], b_3] = \sum_4 A_{1234} b_4 \quad (4)$$

$$[[b_1, b_2^+], b_3^+] = \sum_4 B_{1234} b_4^+ \quad (5)$$

$$[b_1, b_2] = [b_1^+, b_2^+] = 0 , \quad (6)$$

where cumbersome expressions of coefficients functions A_{1234} and B_{1234} and their symmetry properties are not specified here (for them see Ref. 17). Just note that for Frenkel-type excitons A_{1234} and B_{1234} are reduced to $\frac{2}{\mathcal{N}} \delta_{1+3-2,4}$ and $-\frac{2}{\mathcal{N}} \delta_{2+3-1,4}$ (\mathcal{N} is the number of unit cells in the crystal) respectively, and thus our formulae (4) and (5) recover those given in Refs. 15 and 16. The commutation relations (4) to (6) look very like those of the parafermions¹⁸ so that the statistics of excitons is expected to be parafermion-like.

Because of nonbosonity the exciton total number operator \hat{N} proves to be defined not as usually but in a special way as¹⁷

$$\hat{N} = \frac{1}{2\mathcal{N}} \sum_i \{[b_i^+, b_i] + 1\} . \quad (7)$$

Indeed, in Ref. 17 we have proved that if N and $|N\rangle$ are the eigennumber and the eigenstate of the operator \hat{N} then N will get integer values ranging from 0 to \mathcal{N} and $|N\rangle$ will serve as an N -exciton state which is determined in the following form:

$$|N\rangle = C_N (b^+)^N |0\rangle \quad (8)$$

$$C_N = [(\mathcal{N}-1)(\mathcal{N}-2)\dots(\mathcal{N}-N+1)\mathcal{N}^{1-N} N!]^{-1/2} . \quad (9)$$

It is worthy here to notice that Eq. (36) in Ref. 17 was unfortunately misprinted (The quantities in a bracket should be $1 - (N-1)/\mathcal{N}$ but not $(1-N-1)/\mathcal{N}$.)

The above results are general and applicable to both types of excitons: the Frenkel-type in molecular crystals and the Wannier-one in semiconductors. For simplicity and definiteness, in the next sections we shall consider only the former

type of excitons. For the latter type more accurate numerical calculations connected with concrete expressions of functions f_ν need to be performed.

3. Optical Bistability

In this section we shall apply the general commutation relations (4) to (6) to study resonatorless^{19–25} optical bistability in a Frenkel-type exciton system. For clarity, it is divided into several subsections.

3.1. Characteristic equations of optical bistability theory

Following Refs. 2, 15, 16 and 26 a Frenkel-type exciton system subjected to a pumping monochromatic laser field of wavevector \mathbf{k} , real amplitudes A_k and frequency Ω_k can be described by a total Hamiltonian of the form

$$H = \sum_{\mathbf{p}} \left[E_p b_{\mathbf{p}}^+ b_{\mathbf{p}} + (\omega_p + B) c_{\mathbf{p}}^+ c_{\mathbf{p}} + \frac{1}{4} \mathcal{N} F \rho_{\mathbf{p}} \rho_{-\mathbf{p}} + R (b_{\mathbf{p}}^+ c_{\mathbf{p}} + c_{\mathbf{p}}^+ b_{\mathbf{p}}) \right] - \sqrt{\frac{\omega_k}{2}} A_k e^{-i\Omega_k t} c_{\mathbf{k}}^+ + \text{h.c.} \quad (10)$$

$$\rho_{\mathbf{p}} = \rho_{-\mathbf{p}}^+ = \frac{2}{\mathcal{N}} \sum_{\mathbf{q}} b_{\mathbf{p}+\mathbf{q}}^+ b_{\mathbf{q}}. \quad (11)$$

To avoid complicated formulae we confine ourselves only to the consideration of excitons in the lowest state and hence from Eqs. (10) and (11) onwards we shall not write the index ν any more. Then $b_{\mathbf{p}}$ ($c_{\mathbf{p}}$) simply denotes the decay of an exciton (a photon) with quasimomentum \mathbf{p} and quasienergy E_p (frequency ω_p). B comes from A^2 -type of the light-matter interaction Hamiltonian.² F and R stand for the exciton-exciton and exciton-photon coupling constants. While the expressions for B and R can be derived from first principles,² that for F seems not easy to obtain. It, i.e., \sqrt{F} , is most often being introduced phenomenologically to account for all of the possible exciton-exciton interaction mechanisms. Later, in Sec. 4 we will show that F might be determined experimentally by measuring polariton dispersions at different levels of excitation.

Returning now to the commutation relations (4)–(6), we can write the equations of motion for the total number operator of excitons \hat{N} given by Eq. (7) as well as for that of photons $\hat{P} = \sum_{\mathbf{p}} c_{\mathbf{p}}^+ c_{\mathbf{p}}$ (see, e.g., Ref. 26). Solving these equations of motion for the steady state within a mean field approximation gives

$$I = n(1 - n)^{-1} [(n + \alpha)^2 + \beta^2] \quad (12)$$

$$\Phi = n(1 - n)^{-1} [(n + \gamma)^2 + \lambda^2] \quad (13)$$

where I and Φ label the normalized dimensionless input and output light intensities; $n = 2\langle \hat{N} \rangle / \mathcal{N}$ is the relative exciton density in the steady state corresponding to

a given input intensity I ; α , β , λ and λ serve as functions of such parameters as exciton (photon) depopulation and dephase times $\tau_{\parallel b}$ and $\tau_{\perp b}$ ($\tau_{\parallel c}$ and $\tau_{\perp c}$), detuning, etc.^{26,27} Eqs. (12) and (13) are the characteristic equations from which we shall look for the conditions of the appearance of optical bistability in the forthcoming subsection.

3.2. Necessary and sufficient conditions for optical bistability

Optical bistability is understood as a phenomenon when one and the same value of input intensity I results in two stable output intensity values, say, $\Phi = \Phi_1$ and Φ_2 . As seen from Eqs. (12) and (13) Φ seems to have no direct dependence on I . The dependence of Φ on I takes place in fact through the dependence of n on I . If the curve $n = n(I)$ were S-shaped there exists some interval inside which one value of I corresponds to three values of n (among them two are stable but not the third). According to Eq. (13), these values of n in turn give the corresponding values of Φ so that the ($I - \Phi$)-correspondence is one-to-three from the mathematical points of views and one-to-two disregarding the physically unstable Φ -value, and then one says that optical bistability occurs. The conditions for optical bistability to happen, therefore, must be found from the request that the equation $dI/dn = 0$ should possess two distinct roots. These roots must be neither less than zero nor greater than unity, because n loses its physical meaning otherwise (see the definition of n as well as Ref. 28). Using Eq. (12) it follows that $dI/dn = 0$ is equivalent to

$$n^3 + \left(\alpha - \frac{3}{2}\right)n^2 - 2\alpha n - \frac{1}{2}(\alpha^2 + \beta^2) = 0 . \quad (14)$$

Equation (14) is a cubic one with its discriminant being equal to $-108Q$ where²⁹

$$Q = [\beta^2 + (\alpha + 1)^2] \left[\beta^2 - \frac{1}{27}(8\alpha + 9)\alpha^2 \right] . \quad (15)$$

It is well-known that Eq. (14) has three different real roots if $Q < 0$ that means

$$(8\alpha + 9)\alpha^2 > 27\beta^2 . \quad (16)$$

For the three roots to be positive we invoke the so-called Decart sign rule which requires finally

$$0 > \alpha . \quad (17)$$

To show that the conditions (16) and (17) are necessary and at the same time sufficient conditions for optical bistability to appear more detailed analysis should be done. From Eq. (12) we obtain the following asymptotic properties of the curve $I = I(n)$

$$I(0) = 0, \quad I(1 - 0) = \infty \quad (18)$$

$$I(1 + 0) = I(\infty) = -\infty \quad (19)$$

and the second order derivative of I with respect to n

$$\frac{d^2I}{dn^2} = 2(1 - n)^{-3}[(n - 1)^3 + (\alpha + 1)^2 + \beta^2]. \quad (20)$$

Obviously, for $Q < 0$ there are three distinct real roots of Eq. (14) which correspond to either extrema or inflection points of the curve $I = I(n)$. Since $d^2I/dn^2 < 0$ for $n > 1$ (see Eq. (20)), the interval $[1, \infty]$ of n can contain only maxima of the curve $I = I(n)$ but not minima nor inflection points. Paying attention to the properties (19) leads to the fact that one and only one maximum of the curve $I = I(n)$ must exist in the interval $[1, \infty]$. As to the physical interval $[0, 1]$, it cannot contain any inflection points of the curve $I = I(n)$ because $d^2I/dn^2 \neq 0$ for $n \in [0, 1]$ (see again Eq. (20)). Now accounting for the properties (18) we can recognize that the only possible situation is that in which the curve $I = I(n)$ has first a maximum and then a minimum for growing n . This means that the curve $I = I(n)$ has the shape of letter N whereas the curve $n = n(I)$ gets S -shaped. And therefore it can be concluded that the inequalities (16) and (17) are indeed the necessary and sufficient conditions for the occurrence of optical bistability.

3.3. Output-input characteristics shapes

Optical bistability phenomena that occur in a resonator (single³⁰ or double³¹) and possibly with an additional feedback may in general exhibit very complicated output-input characteristics.³⁰⁻³² In this subsection we shall qualitatively show that the characteristics of resonatorless, or intrinsic, optical bistability might also be of various shapes including those producing butterfly-like and three-winged-bow-like hysteresis loops.²⁵

From Sec. 3.2 we know that if conditions (16) and (17) are met optical bistability will take place regardless of the kind of correspondence between Φ and n . Concerning the said correspondence, thanks to the similarity of Eq. (13) in comparison with Eq. (12), i.e. the $(\Phi - n)$ -correspondence, will either be one-to-three or one-to-one depending on whether the following inequalities

$$(8\gamma + 9)\gamma^2 > 27\lambda^2 \quad \text{and} \quad 0 > \gamma \quad (21)$$

simultaneously hold or not. If Conditions (20) were not satisfied, but Conditions (16) and (17) were, then $(\Phi - I)$ -characteristics have S -like shape while $(\Phi - n)$ -ones are monotonically increasing functions (see Fig. 1). When all the Conditions (16), (17) and (20) are met, both curves $\Phi = \Phi(n)$ and $I = I(n)$ get the shape of

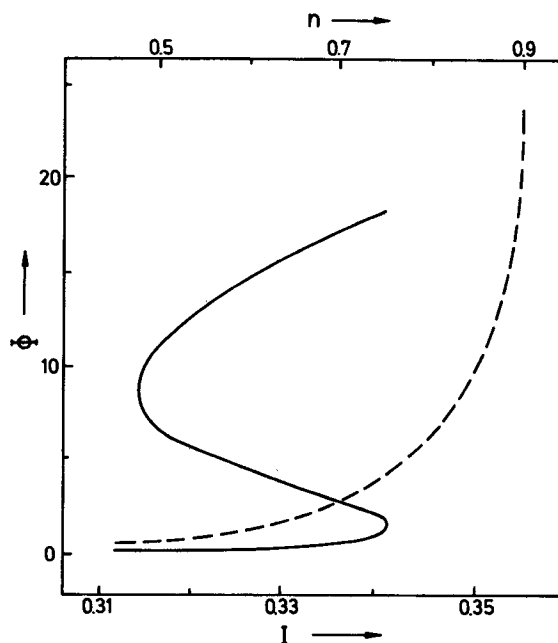


Fig. 1. Φ as a function of I (solid line) and of n (dashed line) for the case of $\alpha = -1.07$, $\beta^2 = 7.3 \cdot 10^{-3}$, $\gamma = 0$ and $\lambda^2 = 0.8$.

letter N and, therefore, the input intensity will dictate the output intensity in delicate manners depending upon many factors. In Ref. 10 the authors drew Φ versus I for some set of parameters and obtained a knot-like form of the curve $\Phi = \Phi(I)$ (see Fig. 2b). Yet, the authors of Ref. 10 did not pay attention to the questions of what other topologically different shapes the curve $\Phi = \Phi(I)$ may have and what factors govern the kind of shapes. Such questions were discussed in Ref. 30 where it was claimed that the topological kind of shapes of the output-input characteristics depends on the sequence of the turning points n_1 , n_2 and n_1^* , n_2^* of the curves $n = n(\Phi)$ and $n = n(I)$. They, however, still omitted some possible situations.^{25,26,30} In Ref. 27 we represented a convenient graphical method to qualitatively analyse different possibilities. It turns out that²⁵ the resonatorless optical bistability curves $\Phi = \Phi(I)$ are affected not only by the successive order of n_1 , n_2 , n_1^* and n_2^* as stated, e.g., in Ref. 30 but also by at least three other factors: (i) the values of the turning points, (ii) the values of extrema of the functions $I = I(n)$ and $\Phi = \Phi(n)$ and (iii) the concrete shapes of the curves $n = n(I)$ and $n = n(\Phi)$. In Fig. 2 we schematically draw 12 possible topologically distinct shapes of the output-input characteristics. Note that only four of them (Figs. 2a, c, e and f) were plotted in Ref. 30 while in Ref. 10 just that in Fig. 2b was drawn.

Now let us look at hysteresis loops (follow arrows) of optical bistability processes. We can observe that the loops in Figs. 2c, d, e, k and l go counter-clockwise

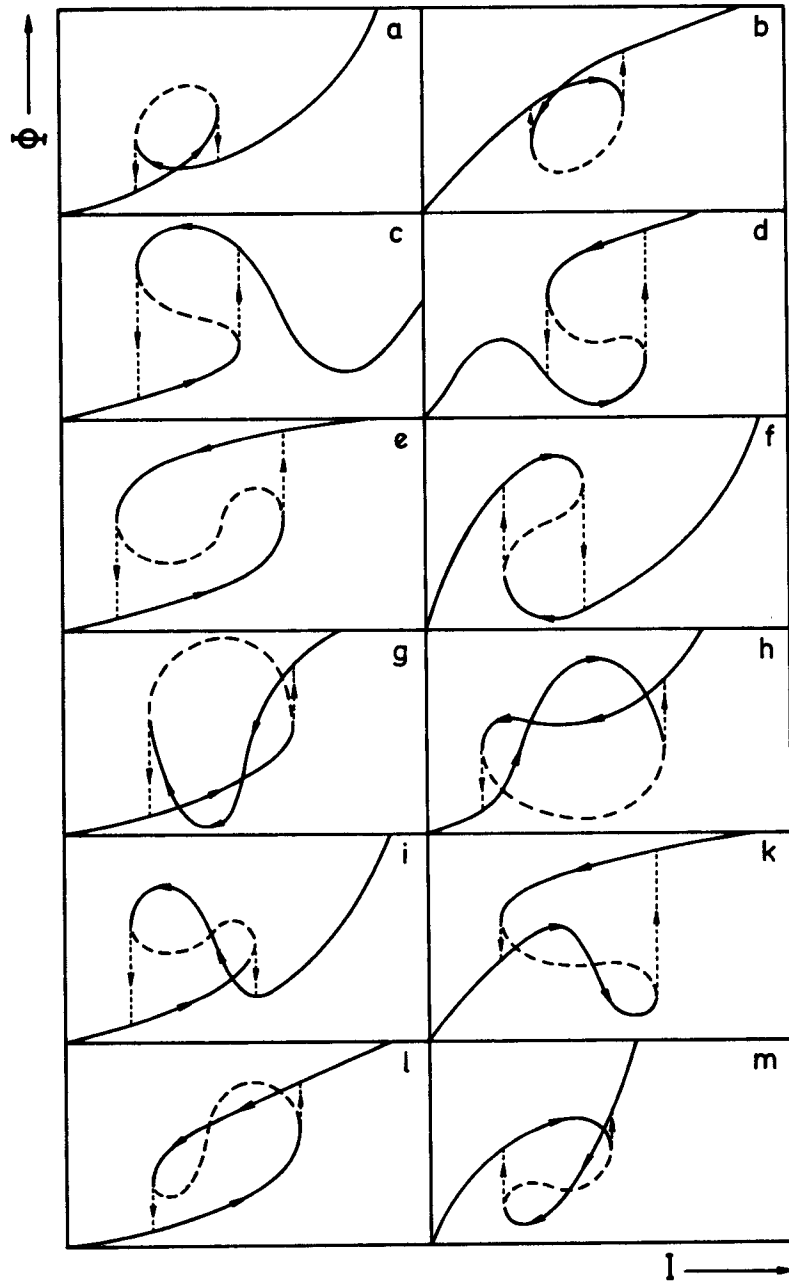


Fig. 2. Possible shapes of the output-input characteristics of resonatorless optical bistability (schematically). Dashed (solid) lines represent unstable (stable) states. Vertical dotted lines show switchings. Directions of hysteresis loops are indicated by arrows.

whereas that in Fig. 2f develops clockwise. Further, the loops in Figs. 2a and c (2b and 2m) have shapes like a butterfly with both switchings down (up). Finally, the loops in Figs. 2g and 2h undergo paths resembling a bow with three wings.²⁵ The variety of output-input characteristics shapes opens a wide chance of, e.g., transforming a cw laser radiation into a pulse sequence,^{33,34} the form of which could be tailored by properly choosing the input parameter and the mechanism generating optical bistability.

4. Nonlinear Polaritons

The concept of polaritons was introduced long ago treating polaritons as intramedium mixed eigenmodes of photons and the medium quasiparticles (phonons, excitons, magnons, . . .). Since they are thought to be real eigenmodes that actually exist in the medium, polariton theory has been developed by many authors and it seems quite successful in interpreting a lot of optical phenomena such as luminescence,^{35,36} absorption,³⁷⁻³⁹ light scattering,⁴⁰⁻⁴² nonlinear effects,^{43,44} additional waves,⁴⁵ etc. The above cited works, nevertheless, considered polaritons without taking into account mutual interactions among quasiparticles in the medium and hence a polariton was understood as a coupled mode between a photon and a quasiparticle and it could be called a linear polariton. Linear polaritons make sense at low excitation level only. In highly photo-excited media a many-quasiparticle system will be established which due to the mutual interaction among quasiparticles can reach a thermal quasiequilibrium under certain conditions. The polariton concept to be used for such a high density system must be retreated to take necessary accounts for the mutual quasiparticle-quasiparticle interaction.⁴⁶⁻⁵² Now not only a photon and a quasiparticle but a number of them should mix to form polaritons which are more correctly referred to as giant⁴⁶ or nonlinear⁴⁸ polaritons. Nonlinear polariton characteristics are found to be dependent on the excitation level. In what follows we shall analyse the explicit dependence of eigenfrequency and damping of nonlinear polaritons on quasiparticle density as well as on exciting laser intensity. As for the latter dependence, the polariton dispersion relations might display some anomalies.⁵¹ The former one, on the other hand, would lead to a situation which could be exploited to experimentally determine the quasiparticle-quasiparticle effective coupling constant.⁵²

4.1. Density-dependent polaritons

Assume that a given monochromatic laser field generates in the medium a coherent many-exciton system which reaches its stationary state. In the stationary state the system can be characterized by a certain stationary exciton density. This density of course is a function of frequency and intensity of the pumping laser field. In this subsection, however, we shall consider it as a given internal parameter and so, for the time being, we can put aside the evidently field-dependent

parts in Eq. (10) in the study of density-dependent polaritons. In the next subsection the dependence of exciton density on field intensity will be discussed, yielding possible anomalies in dispersion relations of nonlinear polaritons.

Define now the following retarded Green functions:

$$G_1(t) = \theta(t)\langle [b(t), b^+(0)] \rangle \quad (22)$$

$$G_2(t) = \theta(t)\langle [c(t), b^+(0)] \rangle \quad (23)$$

$$G_3(t) = \theta(t)\langle [b^+(t)b(t)b(t), b^+(0)] \rangle \quad (24)$$

$$G_4(t) = \theta(t)\langle [b(t)b^+(t)b(t), b^+(0)] \rangle \quad (25)$$

$$G_5(t) = \theta(t)\langle [b^+(t)b(t)c(t), b^+(0)] \rangle \quad (26)$$

where $b(t)$ and $c(t)$ are time-dependent operators in Heisenberg representation, $\langle \dots \rangle$ implies the average over the eigenstate of the field-independent parts of the Hamiltonian (10) and $\theta(t)$ denotes the step function. Using the commutation relations (4)–(6) we are able to determine the time-derivatives of $G_1(t)$ and $G_2(t)$ which are expressed through $G_3(t)$, $G_4(t)$ and $G_5(t)$

$$\begin{aligned} \frac{dG_1(t)}{dt} = & (1 - n)\delta(t) - iEG_1(t) - iEG_2(t) \\ & + \frac{i}{\mathcal{N}}(2E - F)G_3(t) - \frac{iF}{\mathcal{N}}G_4(t) + \frac{2iR}{\mathcal{N}}G_5(t) \end{aligned} \quad (27)$$

$$\frac{dG_2(t)}{dt} = -i(\omega + B)G_2(t) - iRG_1(t) . \quad (28)$$

Transforming (26), (27) into the energy representation gives

$$(\mathcal{E} - E)G_1(\mathcal{E}) = i(1 - n) + RG_2(\mathcal{E}) + \frac{F - 2E}{\mathcal{N}}G_3(\mathcal{E}) + \frac{F}{\mathcal{N}}G_4(\mathcal{E}) - \frac{2R}{\mathcal{N}}G_5(\mathcal{E}) \quad (29)$$

$$(\mathcal{E} - \omega - B)G_2(\mathcal{E}) = RG_1(\mathcal{E}) \quad (30)$$

where \mathcal{E} stands for the energy variable of Green functions $G_i(\mathcal{E})$ in the energy representation. Solving Eqs. (28) and (29) for $G_1(\mathcal{E})$ and $G_2(\mathcal{E})$ within what is known as the random phase approximation⁵¹ yields

$$G_1(\mathcal{E}) = \frac{i(1-n)(\mathcal{E} - \omega - B)}{[\mathcal{N} - (1-n)E - nF](\mathcal{E} - \omega - B) - (1-n)R^2} \quad (31)$$

$$G_2(\mathcal{E}) = \frac{iR(1-n)}{[\mathcal{N} - (1-n)E - nF](\mathcal{E} - \omega - B) - (1-n)R^2} \quad (32)$$

Not incidentally, $G_1(\mathcal{E})$ and $G_2(\mathcal{E})$ have the same denominator. This should be so because both $G_1(\mathcal{E})$ and $G_2(\mathcal{E})$ now represent the dressed quasiparticles — the real exciton-photon coupled eigenmodes (the polaritons) — which actually exist in the medium. The polariton dispersion laws are followed from the common poles of the dressed Green functions $G_1(\mathcal{E})$ and $G_2(\mathcal{E})$ as below

$$\mathcal{E}_{1,2}(k) = \frac{1}{2} \left[E_k + (F - E_k)n + \omega_k + B \mp \sqrt{[E_k + (F - E_k)n - \omega_k - B]^2 + 4R^2(1-n)} \right] \quad (33)$$

The subscripts 1 and 2 of $\mathcal{E}_{1,2}(k)$ indicate the two-branch structure of the polariton dispersion: 1 (2) corresponds to sign “-” (“+”) and describes the so-called lower (upper) polariton branch. Obviously, the n -dependence is evident and thus gives rise to the name of “nonlinear” polaritons. Thanks to the n -dependence we can add n as a natural variable to the notation of the polariton energy which then reads as $\mathcal{E}_{1,2}(k, n)$.

It is worth noticing at this moment that the polariton dispersion is in fact followed from the solution of the equations of motion (28), (29) which in turn are derived from the original Hamiltonian (10) by applying the nonboson commutation relations (4)–(6). In the low density limit the exciton-exciton interaction is negligibly small and one may put $F = 0$ in (10) making the field-independent parts of it to become quadratic with respect to operators b and c . Then the linear polariton dispersion can be easily obtained by diagonalizing such a quadratic Hamiltonian. For high excitation case an attempt was made in Ref. 49 to construct an effective quadratic Hamiltonian in terms of true boson operators so that from it the correct equations of motion (28) and (29) could be reproduced. Then diagonalizing such an effective Hamiltonian would yield the nonlinear polariton dispersion. Regrettably, the effective Hamiltonian found in Ref. 49, being Hermitian, is not self-consistent (Eq. (16b) in Ref. 49 cannot be derived from the effective Hamiltonian as is stated therein). The should-be effective field-independent Hamiltonian giving the needed equations (28), (29) has the form²⁸

$$H_{\text{eff}} = \sum_{\mathbf{p}} \left[[E_{\mathbf{p}} + (F - E_{\mathbf{p}})n] b_{\mathbf{p}}^{\dagger} b_{\mathbf{p}} + (\omega_{\mathbf{p}} + B) c_{\mathbf{p}}^{\dagger} c_{\mathbf{p}} + R[(1-n)b_{\mathbf{p}}^{\dagger} c_{\mathbf{p}} + c_{\mathbf{p}}^{\dagger} b_{\mathbf{p}}] \right] \quad (34)$$

in which, as it differs from (10), operators b and c are both bosonic. We also observe that H_{eff} written in (33) is non-Hermitian and cannot be simply diagonalized by the well-known Bogolubov transformations. However, the diagonalization of such kind of non-Hermitian Hamiltonians can still be performed by the so-called step-by-step Bogolubov transformation method suggested by us in Ref. 53.

Concerning the nonlinear polariton damping, it depends not only on the dampings of excitons and photons but also on the exciton density. Because of lack of space, those interested in its analytical expression are kindly referred to, e.g., Ref. 26.

Coming back now to Eq. (32). There are two reasons causing modification of nonlinear polariton dispersions in comparison with the case of linear polaritons. One comes from the exciton-exciton interaction which renormalizes the exciton self-energy making it to become n -dependent. Denote by \bar{E} the exciton renormalized energy we have

$$\bar{E} = E + (F - E)n \quad (35)$$

and

$$\frac{\partial \bar{E}}{\partial n} = F - E . \quad (36)$$

The other reason is the n -dependence of the effective exciton-photon coupling (saturation effect²⁸) labelled here by \bar{R}

$$\bar{R}^2 = R^2(1 - n) \quad (37)$$

that gives

$$\frac{\partial \bar{R}^2}{\partial n} = -R^2 . \quad (38)$$

In Ref. 47 these two factors were considered separately within a boson approach. Taking simultaneously into account both factors we arrive at the following.⁵² The energy upper-branch of the nonlinear polaritons is always shifted down for any value of k when n increases. The lower-branch, on the other side, behaves quite delicately. For growing n both \bar{E} and \bar{R}^2 decrease (see Eqs. (34) to (37) by paying attention to the realistic fact that $n \leq 1$ and $F < E$). Since \mathcal{E}_1 increases for increasing \bar{E} and for decreasing \bar{R}^2 , there occurs for increasing n a competition between two tendencies one of which lifts the lower-branch while the other shifts it down (for more detail see Ref. 52). It is not difficult to verify that⁵² there exists, independently of n , a certain value of $k = \bar{k} \equiv E - [(F - E)^2 - R^2](E - F)^{-1}$ such that $d\mathcal{E}_1(k, n)/dn > 0$, $= 0$ or < 0 for $k < \bar{k}$, $= \bar{k}$ or $> \bar{k}$, respectively. That

implies that for increasing n the lower-branch $\mathcal{E}_1(k, n)$ is going up in the interval of small values of k : $k < \bar{k}$ (in agreement with Refs. 50 and 51) and going down for $k > \bar{k}$. At $k = \bar{k}$ all the lower-branches with different n should be intersected in a common point, i.e. $\bar{\mathcal{E}}_1 \equiv \mathcal{E}_1(\bar{k}, n_1) = \mathcal{E}_1(\bar{k}, n_2) = \dots$ with $n_1 \neq n_2 \neq \dots$. As at $k = \bar{k}$ $\mathcal{E}_1(\bar{k}, n)$ does not depend on n we can put $n = 1$ and find directly from Eq. (32) an interesting result: $\bar{\mathcal{E}}_1 \equiv F$! Making use of this feature we could propose a possible way of experimental determination of the effective exciton-exciton interaction constant F which is usually introduced in the original Hamiltonian as a phenomenological parameter. Namely, by measuring dispersion curves of the nonlinear polariton energy lower-branch in different stationary states (different n) one can experimentally observe the abovementioned intersection point whose ordinate will give the value of F . All the discussed features are schematically plotted in Fig. 3.

Before proceeding further we note here that the situation in Fig. 3 holds only within the nonboson treatment. The boson case results just in a lowering of both polariton energy branches for increasing n (see e.g. Refs. 47, 49 and 52) in the whole range of k .

4.2. Intensity-dependent polaritons

To analyse the intensity dependence it seems more conveniently to handle Hermitian Hamiltonians rather than non-Hermitian ones. If we request the

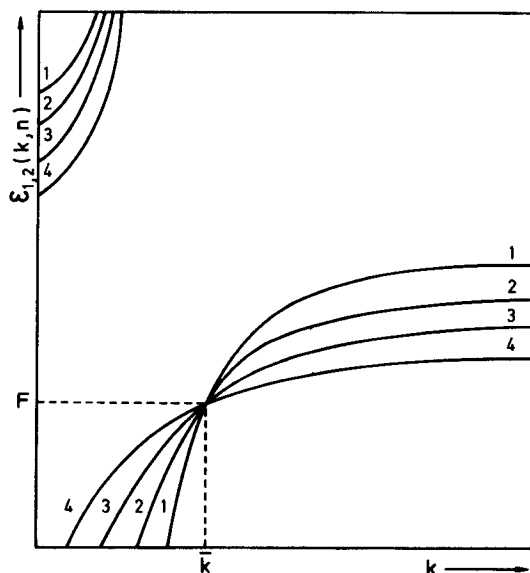


Fig. 3. Schematical dispersion curves of nonlinear polaritons in different stationary states. A curve labelled by a greater number corresponds to a higher excited (larger n) state. All the lower-branch curves are met in a common point whose ordinate equals to F .

effective Hamiltonian to directly produce the polariton dispersion laws (32) but not the system of Eqs. (28), (29), then instead of (33) we shall have another effective Hamiltonian \bar{H}_{eff} which is in this case a Hermitian one

$$\bar{H}_{\text{eff}} = \sum_{\mathbf{p}} [\bar{E}_{\mathbf{p}} b_{\mathbf{p}}^{\dagger} b_{\mathbf{p}} + (\omega_{\mathbf{p}} + B) c_{\mathbf{p}}^{\dagger} c_{\mathbf{p}} + \bar{R} (b_{\mathbf{p}}^{\dagger} c_{\mathbf{p}} + c_{\mathbf{p}}^{\dagger} b_{\mathbf{p}})] \quad (39)$$

where \bar{E} is defined by Eq. (34) and $\bar{R} = R \sqrt{1-n}$. As can be checked, diagonalizing (38) indeed yields the laws (32). The total effective Hamiltonian $\bar{H}_{\text{eff}}^{\text{total}}$ describing a system of coherent excitons and photons that are generated by an external monochromatic laser field has in a rotating frame approximation the below-written form

$$\bar{H}_{\text{eff}}^{\text{total}} = (\bar{E} - \Omega) b^{\dagger} b + (\omega + B - \Omega) c^{\dagger} c + \bar{R} (b^{\dagger} c + c^{\dagger} b) + \bar{A} (c^{\dagger} + c) \quad (40)$$

here $\bar{A} = -(\omega/2)^{1/2} A$ and all the subindices of operators are omitted for brevity.

By means of a unitary transformation and using the concept of averaged values over the coherent state⁵⁴ we can derive from (39) the relation connecting exciton density n with laser intensity I_L which reads

$$I_L = \vartheta n(1-n)^{-1} (n + \xi)^2 \quad (41)$$

with

$$\vartheta = \frac{\mathcal{N} E^2 [4y(x-1) + z^2]}{\omega z^2} \quad (42)$$

and

$$\xi = \frac{4y \left(1 - \frac{\omega}{E}\right) - z^2}{4y(x-1) + z^2} \quad (43)$$

where

$$x = \frac{F}{E}, \quad y = \frac{B}{E} \quad \text{and} \quad z = \frac{2R}{E}. \quad (44)$$

If we denote $I_L \vartheta^{-1}$ by I we observe that Eq. (40) is nothing else but a simplified form of the general equation (12) in which α plays the role of ξ and $\beta = 0$. According to the results reported in Sec. 3.2 one value of laser intensity will correspond to three values (two stable and the third not) of exciton density if the

following inequalities are fulfilled

$$0 > 8\xi > -9 . \tag{45}$$

Combining Eq. (32) and Eq. (40) leads to the fact that polariton dispersion depends on laser intensity parametrically via the intensity-dependence of exciton density. Since for fixed I there might in general be more than one value of n , say, $n = n_1, n_2, \dots$. So, inserting $n = n_i$; into Eq. (32) makes this equation to be “multivalued”, too,

$$\mathcal{E}_{1,2}(k, n) \rightarrow \mathcal{E}_{1,2}(k, n_i), \quad i = 1, 2, \dots \tag{46}$$

and we would get anomalies (see later) in polariton dispersion curves. Such an idea of predicting various anomalies was spoken out for the first time in Ref. 55 and it fairly well explained some anomalous effects observed by experiments on absorption and resonance scattering under intense laser field.^{56,57} Till now, however, the polariton dispersion multistability has been considered, to our knowledge, only by us.^{51,58} For illustration, we draw in Fig. 4 $\bar{\mathcal{E}}_{1,2} = \mathcal{E}_{1,2}/E$ as functions of I for three fixed values of ω/E . For ω/E as much as $0 < \omega/E < 0.75$ the conditions (44) are satisfied and both polariton energy branches fold

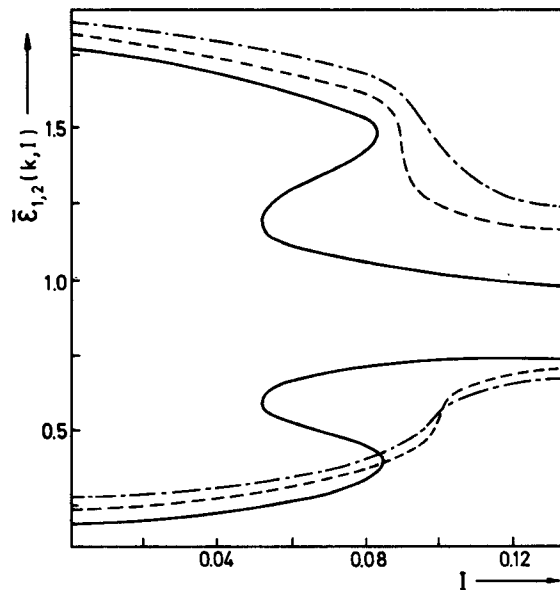


Fig. 4 Polariton dispersion curves as functions of laser intensity for $\omega/E = 0.6$ (full lines), 0.754 (dashed lines) and 0.8 (dashed-dotted lines).

themselves (the upper branch is folded down, while the lower one does the opposite) to generate anomaly as seen from the full lines in Fig. 4.

To finish this subsection we would like to say some words regarding the great complexity of the shape of polariton dispersion curves. If we account for the photon-photon interaction the polariton energy depends also on intramedium-photon density n^{ph}

$$\mathcal{E}_{1,2} \rightarrow \mathcal{E}_{1,2}(k, n, n^{\text{ph}}) \quad (47)$$

(see, e.g., Eq. (27) in Ref. 51). The photon density, in its turn, depends on exciton density like I_L does (see Eq. (43) in Ref. 51 and Eq. (40) in this work). Judging similarly as we did in Sec. 3 for Eqs. (12) and (13) it is expected that the way the photon density depends on the laser intensity must be very complicated (see for comparison the dependence of Φ on I in Sec. 3). The intensity-dependence of polariton dispersion curves thus takes place through the dependence of both n and n^{ph} on I . One hardly can numerically plot all possible shapes of the curves and as a rule a thorough study will be needed from a topological point of view.

5. Concluding Remarks

More and more nature forces scientists to go far beyond the traditional looks at it. Now not only bosons and fermions in three-dimension space but also anyons in space dimension two, not only integer and neutral charge but also fractional one, not only crystal and liquid but also quasicrystal, not only order and disorder but also chaos, . . . all these require a specific and at the same time unified method for their consideration. The nonboson treatment in this article though being a good extension of that developed in Refs. 15 and 16 is still far from completely reliable. The theoretical predictions (see text) of this treatment need to be confirmed experimentally. However, due to the importance of the strongly excited materials physics towards practical application and the missing of fully rigorous theory for studying high density systems of composite particles, the presented treatment here can serve as a fair tool to deal with many-exciton systems and even with systems containing biexcitons and impurities. Large optical nonlinearity has been observed due to two-photon resonance of the biexciton.⁵⁹ Formation of biexcitons in semiconductors may give rise to the appearance of optical bistability⁵⁹⁻⁶⁹ that up to now is studied just in the approximation in which biexcitons are taken as ideal bosons. Comprising four elementary particles, two electrons and two holes, biexcitons of course manifest larger nonbosonity than excitons do. Further, at strong excitation level excitons and biexcitons are no longer independent quasiparticles: their operators do not commute with each other^{70,71} and thus handling high density exciton-biexciton systems is a highly challenging problem to be resolved. It is hoped that the nonboson treatment might be somehow helpful in finding closed commutation

relations for both exciton and biexciton operators separately as well as commonly. We shall try to do this in the future.

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