Quantum Optical Effects and Nonlinear Dynamics in Interacting Electron Systems

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(Received 16 July 1996)

We present a theory of the nonlinear optical response of cavity embedded interacting electron systems which does not rely on semiclassical factorization. The theory provides a unified basis for understanding nonlinear quantum dynamics in cavity embedded quantum wells. Nonclassical behavior of transmitted light is found. [S0031-9007(96)01797-8]

PACS numbers: 42.50.–p, 42.65.–k, 71.35.–y, 71.36.+c

The atom-cavity system has been used to investigate quantum dynamical processes for open quantum systems in a regime of strong coupling and to explore quantum behaviors that have no classical counterparts [1]. With the development of crystal growth technologies, it has become possible to investigate cavity embedded interacting electron systems [2–6]. Cavity quantum electrodynamics (cavity QED) effects have been observed by placing quantum wells (QW's) in a semiconductor planar microcavity [2,7]. The QW exciton-cavity system has some fundamental differences with respect to the much simpler two level atom-single mode cavity system; however, coherent linear dynamics of the two systems is very similar despite the complexity of the electronic states of the semiconductor. In fact, a weak light beam of given wave vector can excite only one-exciton states, and, owing to the conservation of the in-plane momentum **k**, only the exciton state with the same wave vector of incident light interacts. A situation similar to linear dynamics of two level atoms in a single mode microcavity is thus reproduced. Nonlinear dynamics does not maintain the simple picture of two coupled fields. Furthermore the source of nonlinearities in two level atoms comes from saturation, while excitonic nonlinearity comes also from Coulomb interaction between electrons. As a consequence a more complex situation and new phenomena are expected exploring nonlinear dynamics of interacting electron systems in the strong coupling regime. Recently a first principle semiclassical theory of nonlinear response for a two band semiconductor model has been given in [8,9]. The theory, applied to four-wave mixing in quantum wells, provides a unified basis for understanding a wide range of observed phenomena. Quantum optical effects and manifestation of nonclassical dynamics have been predicted [10,11] and observed [12,13] experimentally in atom-cavity systems also for a large number of atoms. The nonlinear behavior of the strongly coupled exciton-cavity system [14] opens a new and versatile way of exploring quantum dynamics in mesoscopic systems. Time resolved photon statistics during normal mode coupling in a semiconductor microcavity has been recently measured [15].

A full quantum mechanical description of light in interaction with a confined polarizable medium, in the linear regime, has been given in [16]. Here we present a theory of the nonlinear optical response of an interacting electron system which does not rely on the semiclassical factorization. This opens a promising connection between the physics of collective excitations and quantum optics. The theory provides both information on quantum correlations of emitted photons, predicting quantum optical effects, and a first principle description of those well-known nonlinear phenomena in exciton physics as four-wave mixing and hyper-Raman scattering. The system which we investigate consists of a quantum well (QW) grown inside a semiconductor planar Fabry-Perot resonator. We treat the cavity field within the quasimode approximation, the cavity field is quantized as though the mirrors where perfect, and the resulting discrete modes are coupled phenomenologically to the external continuum of modes [17]. The eigenstates of the Hamiltonian H_{ph} of the cavity modes are written as $|n, \lambda\rangle$, where n indicates the total number of photons in the state and $\lambda = (\mathbf{k}_1, \sigma_1; \ldots; \mathbf{k}_n, \sigma_n)$ is a label specifying wave vector $\mathbf{k}$ and polarization σ of each of the n photons. The states $|E_{N,\alpha}\rangle$ with energy $\omega_{N,\alpha}$ of the Hamiltonian H_c of the usual semiconductor model can be labeled according to the number N of *eh* pairs [9,18]. The state $|E_{N=0}\rangle$ is the semiconductor ground state. The $N = 1$ subspace is the exciton subspace with the additional quantum number $\alpha = (n, \sigma, \mathbf{k})$. The set of states with $N = 2$ *eh* pairs determines the biexciton subspace. The interaction of the electron system with cavity modes is given in the usual rotating wave approximation by

$$H_I = i\hbar \sum_{n,k} V^*_{n,k} a^\dagger_k B_{n,k} + \text{H.c.}, \qquad (1)$$

where the operator $a^\dagger_k$ creates a photon state with $k \equiv (\mathbf{k}, \sigma)$ and energy $\omega_k = (\omega_0^2 + v^2\mathbf{k}^2)^{1/2}$, v being the velocity of light inside the cavity, $B^\dagger_{n,k}$ creates an exciton state with the same wave vector and polarization k and energy $\omega_{1,n,k}$. $V_{n,k}$ is the photon-exciton coupling coefficient enhanced by the presence of the cavity [19]. The dephasing of the semiconductor excitations is introduced phenomenologically, by assuming a reservoir at zero temperature, while the linear coupling of cavity modes with the external modes provides both the damping and the

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input optical pumping of the cavity modes [17]. For a coherent input beam, the driving of cavity modes is described by the following Hamiltonian term

$$H_p = \sum_k \mathcal{E}_k (a_k^\dagger - a_k), \tag{2}$$

where $\mathcal{E}_k$ are the amplitudes of the coherent driving fields. The dynamical evolution of the coupled system is governed by the expectation values of the following product Hubbard operators: $\hat{X}_{N,\alpha;M,\beta}\hat{Y}_{n,\lambda;m,\mu}$, where $\hat{X}_{N,\alpha;M,\beta} = |E_{N,\alpha}\rangle\langle E_{M,\beta}|$, $\hat{Y}_{n,\lambda;m,\mu} = |n,\lambda\rangle\langle m,\mu|$. Hubbard operators can be used to express the exciton [9] and photon operators. From the form of the interaction and of the pump Hamiltonian, the expectation values of these operators can be expressed as a power series in the external field

$$\langle \hat{X}_{N,\alpha;M,\beta}\hat{Y}_{n,\lambda;m,\mu}\rangle = \sum_{i=0}^{i_0} \langle \hat{X}_{N,\alpha;M,\beta}\hat{Y}_{n,\lambda;m,\mu}\rangle^{(N+M+n+m+2i)} + O(\mathcal{E}^{N+M+n+m+2i_0+2}). \tag{3}$$

Equation (3) is of great relevance for the following calculations, since it provides a truncation scheme based on powers of excitation strength. A similar truncation scheme, in the semiclassical framework, was first exploited by Axt and Stahl [20], and Victor, Axt, and Stahl [21]. The state of the quantum system is defined by a density operator and a master equation describing its time evolution. The full analytic solution of such an equation is not known even for the much simpler two level atoms-single mode cavity system [11]. Instead of solving the master equation, we consider the equation of motion for the expectation values of system operators of interest, up to a given order. We start by considering the polariton linear dynamics writing the coupled equation of motion for $\langle a_k\rangle^{(1)} = \langle \hat{X}_{0;0}\hat{Y}_{0;1,k}\rangle^{(1)}$ and $\langle B_{n,k}\rangle^{(1)} = \langle \hat{X}_{0;n,k}\hat{Y}_{0;0}\rangle^{(1)}$. We obtain

$$\frac{\partial}{\partial t}\langle a_k\rangle^{(1)} = -\gamma_k'\langle a_k\rangle^{(1)} + \sum_n V_{n,k}\langle B_{n,k}\rangle^{(1)} + \mathcal{E}_k, \tag{4a}$$

$$\frac{\partial}{\partial t}\langle B_{n,k}\rangle^{(1)} = -\Gamma_{n,k}'\langle B_{n,k}\rangle^{(1)} - V_{n,k}\langle a_k\rangle^{(1)}. \tag{4b}$$

In these equations $\gamma_k' = \gamma + i\omega_k$, where γ is the cavity damping, assumed for simplicity independent on the mode, and analogously $\Gamma_{n,k}' = \Gamma + i\omega_{1,n,k}$. In the following we will show as a unified description of the lowest order nonlinear quantum dynamics of the system can be obtained in terms of the solutions of linear equations (4) and of the two particle correlation functions $\langle a_k a_{\tilde{k}}\rangle^{(2)}$, $\langle B_{n,k} a_{\tilde{k}}\rangle^{(2)}$, and $\langle B_{\tilde{n},\tilde{k}} B_{n,k}\rangle^{(2)}$, obeying the following set of coupled equations

$$\frac{\partial}{\partial t}\langle a_k a_{\tilde{k}}\rangle^{(2)} = -(\gamma_k' + \gamma_{\tilde{k}}')\langle a_k a_{\tilde{k}}\rangle^{(2)} + \sum_n (V_{n,\tilde{k}}^*\langle a_k B_{n,\tilde{k}}\rangle^{(2)} + V_{n,k}^*\langle a_{\tilde{k}} B_{n,k}\rangle^{(2)}) + \mathcal{E}_{\tilde{k}}\langle a_k\rangle^{(1)} + \mathcal{E}_k\langle a_{\tilde{k}}\rangle^{(1)}, \tag{5a}$$

$$\frac{\partial}{\partial t}\langle a_{\tilde{k}} B_{n,k}\rangle^{(2)} = -(\Gamma_{n,k}' + \gamma_{\tilde{k}}')\langle a_{\tilde{k}} B_{n,k}\rangle^{(2)} + \mathcal{E}_{\tilde{k}}\langle B_{n,k}\rangle^{(1)} - V_{n,k}\langle a_k a_{\tilde{k}}\rangle^{(2)} + \sum_{\tilde{n}} V_{\tilde{n},\tilde{k}}^*\langle B_{\tilde{n},\tilde{k}} B_{n,k}\rangle^{(2)}, \tag{5b}$$

$$\frac{\partial}{\partial t}\langle B_{\tilde{n},\tilde{k}} B_{n,k}\rangle^{(2)} = -(\Gamma_{n,k}' + \Gamma_{\tilde{n},\tilde{k}}')\langle B_{\tilde{n},\tilde{k}} B_{n,k}\rangle^{(2)} - V_{\tilde{n},\tilde{k}}\langle a_{\tilde{k}} B_{n,k}\rangle^{(2)} - V_{n,k}\langle a_k B_{\tilde{n},\tilde{k}}\rangle^{(2)} + R_{n,k;\tilde{n},\tilde{k}}^{(2)}, \tag{5c}$$

where $R_{n,k;\tilde{n},\tilde{k}}^{(2)}$ is given by

$$R_{n,k;\tilde{n},\tilde{k}}^{(2)} = \sum_{k',\beta} \Omega_{n,k;\tilde{n},\tilde{k};k';\beta}^{(1)}\langle a_{k'} B_\beta\rangle^{(2)} - i\sum_\beta c_{n,k;\tilde{n},\tilde{k};\beta}^{(1)}\langle \hat{X}_{0;2,\beta}\rangle^{(2)}, \tag{6}$$

with

$$c_{n,k;\tilde{n},\tilde{k};\beta}^{(1)} = (\omega_{2,\beta} - \omega_{1,n,k} - \omega_{1,\tilde{n},\tilde{k}})\langle E_{1,\tilde{n},\tilde{k}}|B_{n,k}|E_{2,\beta}\rangle, \tag{7a}$$

$$\Omega_{n,k;\tilde{n},\tilde{k},k';\beta}^{(1)} = -\sum_{n'} V_{n',k'}\langle E_{1,\tilde{n},\tilde{k}}|[B_{n,k}, B_{n',k'}^\dagger] - \delta_{(n,k);(n',k')}|E_{1,\beta}\rangle. \tag{7b}$$

The equation of motion for $\langle \hat{X}_{0;2,\beta}\rangle^{(2)}$ reads

$$\frac{\partial}{\partial t}\langle \hat{X}_{0;2,\beta}\rangle^{(2)} = -(2\Gamma + i\omega_{2,\beta})\langle \hat{X}_{0;2,\beta}\rangle^{(2)} - \sum_{n',k';n'',k''} V_{n'',k''}\langle E_{2,\beta}|B_{n',k'}|E_{1,n'',k''}\rangle\langle a_{k'} B_{n'',k''}\rangle^{(2)}. \tag{8}$$

We stress that all deviations from semiclassical results in the two particle correlation functions are determined by $R_{n,k;\tilde{n},\tilde{k}}^{(2)}$; if it would be zero only trivial linear and semiclassical results would be obtained, i.e., $\langle a_k, a_{\tilde{k}}\rangle^{(2)} = \langle a_k a_{\tilde{k}}\rangle^{(2)} - \langle a_k\rangle^{(1)}\langle a_{\tilde{k}}\rangle^{(1)} = 0$. $R_{n,k;\tilde{n},\tilde{k}}^{(2)}$ can be considered a two exciton correlation force. The coefficients (7) coincide with those obtained in the semiclassical theory of nonlinear response in interacting electron systems [9], and $R_{n,k;\tilde{n},\tilde{k}}^{(2)}$ differs from the factor in the source term for the third order nonlinear response in [9] by semiclassical factorization: if we make the replacement $\langle a_{k'} B_{n'',k''}\rangle^{(2)} \to \langle a_{k'}\rangle^{(1)}\langle B_{n'',k''}\rangle^{(1)}$ in Eqs. (6) and (8), coincidence is achieved. So we can conclude that the same interaction processes and source of nonlinearities determines both semiclassical nonlinear response and quantum correlations. In particular, the first term in Eq. (6) is the phase space filling term, while the second term describes exciton-exciton interaction and biexciton effects [9]. The difference outlined above between the correlation force (6) in the quantum theory, and in the semiclassical approach is important. In fact, in the semiclassical theory the correlation force is a known driving term, since it can be expressed in terms of the coefficients (7) and of the linear response solutions. In the quantum theory the correlation force depends on the two particle correlation functions $\langle a_k B_{n,k'}\rangle^{(2)}$ that it affects. As a consequence a self consistent calculation is required in order to solve Eq. (5). Equation (5) can be interpreted as the extension to interacting electron systems of Eq. (4) of Carmichael *et al.* [11], describing quantum dynamics of N two level atoms in a single mode cavity. We stress the importance of the exciton-exciton correlation force and of two particle correlations for the nonlinear quantum dynamics of the system. In the following we consider some examples which show how nonlinear quantum dynamics can be described in terms of two particle correlation functions.

Four-wave mixing.—If the system evolves as a pure state, the expectation values of Hubbard operators can be written as

$$\langle \hat{X}_{N,\alpha;M,\beta}\hat{Y}_{n,\lambda;m,\mu}\rangle = \langle \hat{X}_{0;N,\alpha}\hat{Y}_{0;n,\lambda}\rangle^* \times \langle \hat{X}_{0;M,\beta}\hat{Y}_{0;m,\mu}\rangle / \langle \hat{X}_{0;0}\hat{Y}_{0;0}\rangle . \quad (9)$$

By using this expression and Eq. (3) we obtain the dependence of third order polarization on two particle correlations in a simple form. Third order polarization is proportional to $\langle B_{n,k}\rangle^{(3)}$. Polariton dynamics implies that the exciton field generated by a nonlinear source term propagates exchanging energy with the cavity field. The coupled propagation up to the third order is described by the following equations:

$$\frac{\partial}{\partial t}\langle a_k\rangle = -\gamma_k'\langle a_k\rangle + \sum_n V_{n,k}\langle B_{n,k}\rangle + \mathcal{E}_k , \quad (10a)$$

$$\frac{\partial}{\partial t}\langle B_{n,k}\rangle = -\Gamma_{n,k}'\langle B_{n,k}\rangle - V_{n,k}\langle a_k\rangle + \sum_{\tilde{n},\tilde{k}}\langle B_{\tilde{n},\tilde{k}}\rangle^{(1)*} R^{(2)}_{n,k;\tilde{n},\tilde{k}} . \quad (10b)$$

The nonlinear source term in Eq. (10b) depends on the correlation force and hence on two particle correlations. This coupled propagation of the nonlinear excitation leads to the coherent oscillations in four-wave mixing observed experimentally [14]. The nonlinear source term in Eq. (10b) coincides with the corresponding term in the semiclassical theory [9] if quantum correlations are neglected. However, we can notice that four-wave mixing is generated by the same correlation force determining two particle quantum correlations; as a consequence, deviations from semiclassical results and renormalization effects [22] are expected.

Hyper-Raman scattering.—The hyper-Raman process can be schematically described as follows: two incident photons of given wave vector $\mathbf{k}_1$, $\mathbf{k}_2$ drive the cavity modes via the Hamiltonian term (2) which, interacting with the electron system, create a two electron-hole pair state which annihilates to create a one electron-hole pair state and one photon of different wave vectors $\mathbf{k}$ and $\tilde{\mathbf{k}}$ such that $\mathbf{k} + \tilde{\mathbf{k}} = \mathbf{k}_1 + \mathbf{k}_2$ [23]. In contrast to the four-wave mixing, this process can be described only by a full quantum mechanical approach, since the last step in the above description, i.e., the spontaneous breaking of the two electron-hole pair state is not allowed in the semiclassical theory. The correlation force drives the process, generating two particle correlations, with wave vectors $\mathbf{k}$, $\tilde{\mathbf{k}}$ such that $\mathbf{k} + \tilde{\mathbf{k}} = \mathbf{k}_1 + \mathbf{k}_2$, which determine light emission in the $\mathbf{k}$ and $\tilde{\mathbf{k}}$ directions. The hyper-Raman spectrum is given by the inelastic part of the spectrum of the fluorescent light,

$$T_{\text{out}}(\omega) = 2\gamma_{\text{out}}\int d\tau\, e^{i\omega\tau}\langle a_k^\dagger(t), a_k(t+\tau)\rangle_{ss} , \quad (11)$$

where the subscript ss indicates steady state and γ_{out} is the loss coefficient of the output mirror, for a symmetric cavity $\gamma_{\text{out}} = \gamma/2$. According to the quantum regression theorem, two time correlation functions as $\langle a_k^\dagger(t), a_k(t+\tau)\rangle$ obey the same equations as $\langle a_k(\tau)\rangle$, with initial conditions given by the equal time correlation functions $\langle a_k^\dagger, a_k\rangle$ and $\langle a_k^\dagger, B_{n,k}\rangle$. By expanding equal time correlation functions in terms of Hubbard operators and using Eqs. (3) and (9), we obtain

$$\langle a_k^\dagger, a_k\rangle^{(4)} = \sum_{\tilde{\sigma}} |\langle a_k, a_{\tilde{k}}\rangle^{(2)}|^2 + \sum_{\tilde{n},\tilde{\sigma}} |\langle a_k, B_{\tilde{n},\tilde{k}}\rangle^{(2)}|^2, \quad (12a)$$

$$\langle a_k^\dagger, B_{n,k}\rangle^{(4)} = \sum_{\tilde{\sigma}}\langle a_k^\dagger, a_{\tilde{k}}^\dagger\rangle^{(2)}\langle a_{\tilde{k}}, B_{n,k}\rangle^{(2)} + \sum_{\tilde{n},\tilde{\sigma}}\langle a_k^\dagger, B_{\tilde{n},\tilde{k}}^\dagger\rangle^{(2)}\langle B_{\tilde{n},\tilde{k}}, B_{n,k}\rangle^{(2)}, \quad (12b)$$

where $\tilde{\mathbf{k}} = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}$. The integrated hyper-Raman emission is proportional to expression (12a). We notice that, to calculate the spectrum at fourth order (second order in the incident intensities) by the quantum regression theorem, it is sufficient to consider the equations for $\langle a_k(\tau)\rangle$ and $\langle B_{n,k}(\tau)\rangle$ up to the third order [Eq. (10)]. Resonances and quantitative evaluations of such spectra will be given in a forthcoming paper; here we have shown the relation between hyper-Raman emission and two particle correlation functions, solution of Eq. (5).

Squeezing.—The exciton-exciton correlation force can induce quantum correlations between photons, leading to nonclassical effects. Let us consider, as an example, the output spectrum of squeezing for an incident beam at normal incidence $\mathbf{k} = 0$ and of given polarization (i.e., +), with energy $\omega_p = \omega_{\mathbf{k}=0}$. We also consider light emission along the same direction and with the same polarization of incident light. The output spectrum of squeezing is proportional to the Fourier transform of the time-ordered and normally ordered correlation function [24] $T\langle : A_1(\tau), A_1 :\rangle_{ss}$, where A_1 is a quadrature component, which we choose in phase with the driving field: $A_1 = a_0 + a_0^\dagger$. We consider a cavity at zero detuning $\omega_{1,n=1} = \omega_0$, where $n = 1$ indicates the heavy-hole $1S$ exciton level, and present a simple analytical approximation avoiding self consistency. If deviations from semiclassical results are not large, we can solve Eq. (5) by considering the correlation force as a known term, making in $R^{(2)}_{n,k;\tilde{n},\tilde{k}}$ the replacement $\langle a_{k'} B_{n'',k''}\rangle^{(2)} \to \langle a_{k'}\rangle^{(1)} \langle B_{n'',k''}\rangle^{(1)}$. Furthermore, since only a (+) polarized incident beam is considered, bound two electron-hole pair states cannot be created, and we can neglect the force-force correlation function [second term in Eq. (10) of [9]]. By assuming $\omega_{1,n\neq 1} - \omega_p \gg \Gamma, \gamma$ we obtain up to the first nonzero order

$$\langle : A_1(0), A_1(0) :\rangle_{ss}^{(2)} = \langle a_0, a_0\rangle_{ss}^{(2)} + \text{c.c.} = -2\,\frac{\Gamma}{\gamma + \Gamma}\,\frac{V_1^2}{\gamma\Gamma + V_1^2} \times \left(\frac{N}{2N_{psf}} + \frac{N}{N_{\text{exch}}}\right). \tag{13}$$

In this expression, showing a noise reduction of A_1 below the noise level of a coherent state, N/N_{psf} and N/N_{exch} depend on the density of excitons generated by the pump beam and coincide with the intensity dependent corrections to the exciton susceptibility [25]. If Coulomb interaction goes to zero $1/N_{\text{exch}}$ becomes zero and (13) coincides with the corresponding expression for the N atoms-cavity system obtained in [26] in the limit of weak driving fields. According to the quantum regression theorem, in order to calculate the spectrum of squeezing at second order, one has to solve linear Eq. (4) with initial conditions given by $\langle a_0, a_0\rangle_{ss}^{(2)}$ and $\langle B_{n,0}, a_0\rangle_{ss}^{(2)}$. The output spectrum of squeezing can be easily obtained by adiabatic elimination of the variables corresponding to out of resonance exciton levels $n \neq 1$. It reads

$$S_{\text{out}}(\omega)^{(2)} = \gamma\langle : A_1(0), A_1(0) :\rangle_{ss}^{(2)}\,\frac{\bar{\Gamma}}{\bar{\Omega}} \times \left[\frac{2\bar{\Omega} - \omega}{\bar{\Gamma}^2 + (\omega - \bar{\Omega})^2} + \frac{2\bar{\Omega} + \omega}{\bar{\Gamma}^2 + (\omega + \bar{\Omega})^2}\right], \tag{14}$$

where $2\bar{\Omega}$ is the Rabi splitting of the exciton-cavity system and $\bar{\Gamma} = (\Gamma + \gamma)/2$ is the normal modes linewidth. We expect this effect to not be easy to be observed experimentally, since noise from thermalized electrons and holes can destroy squeezing. However, for systems with $V_1/(\gamma + \Gamma) \gg 1$, the Rabi peaks are well separated and the energy of the incident beam, between the Rabi peaks at $\omega_p = \omega_{1,1} = \omega_0$, is out of the Rabi splitted resonances, hence, no real population is generated and thermalization effects can be strongly lowered; in addition, squeezing increases by increasing $V_1/(\gamma + \Gamma)$ if the density of excitons N is maintained constant by adjusting the intensity of the incident beam [see Eq. (13)].

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