Dynamics-Controlled Truncation Scheme for Quantum Optics and Nonlinear Dynamics in Semiconductor Microcavities

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Abstract

We present a systematic theory of Coulomb-induced correlation effects in the nonlinear optical processes within the strong-coupling regime. In this paper we shall set a dynamics controlled truncation scheme [1] microscopic treatment of nonlinear parametric processes in SMCs including the electromagnetic field quantization. It represents the starting point for the microscopic approach to quantum optics experiments in the strong coupling regime without any assumption on the quantum statistics of electronic excitations (excitons) involved. We exploit a previous technique, used in the semiclassical context, which, once applied to four-wave mixing in quantum wells, allowed to understand a wide range of observed phenomena [2]. We end up with dynamical equations for exciton and photon operators which extend the usual semiclassical description of Coulomb interaction effects, in terms of a mean-field term plus a genuine non-instantaneous four-particle correlation, to quantum optical effects.

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I. INTRODUCTION

Since the early Seventies [3] researchers have been exploring the possible realization of semiconductor-based heterostructures, devised according to the principles of quantum mechanics. The development of sophisticated growth techniques started a revolution in semiconductor physics, determined by the possibility of confining electrons in practical structures. In addition, the increasing ability in controlling fabrication processes has enabled the manipulation of the interaction between light and semiconductors by engineering, in addition to the electronic wave functions, the light modes.

Entanglement is one of the key features of quantum information and communication technology [4] and a hot topic in quantum optics too. Parametric down-conversion is the most frequently used method to generate highly entangled pairs of photons for quantum-optics applications, such as quantum cryptography and quantum teleportation. Rapid development in the field of quantum information requires monolithic, compact sources of nonclassical photon states enabling efficient coupling into optical fibres and possibly electrical injection. Semiconductor-based sources of entangled photons would therefore be advantageous for practical quantum technologies. The strong light-matter interaction in these systems gives rise to cavity polaritons which are hybrid quasiparticles consisting of a superposition of cavity photons and quantum well (QW) excitons [5]. Demonstrations of parametric amplification and parametric emission in semiconductor microcavities (SMCs) with embedded QWs [6, 7, 8], together with the possibility of ultrafast optical manipulation and ease of integration of these microdevices, have increased the interest on the possible realization of nonclassical cavity-polariton states [9, 10, 11, 12, 13]. In 2004 squeezed light generation in SMCs in the strong coupling regime has been demonstrated [14]. In 2005 an experiment probing quantum correlations of (parametrically emitted) cavity polaritons by exploiting quantum complementarity has been proposed and realized [11]. Specifically, it has been shown that polaritons in two distinct idler modes interfere if and only if they share the same signal mode so that which-way information cannot be gathered, according to Bohr’s quantum complementarity principle.

Laser spectroscopy in semiconductors and in semiconductor quantum structures has been greatly used because exciting with ultrashort optical pulses in general results in the creation of coherent superpositions of many-particle states. Thus it constitutes a very promising
powerful tool for the study of correlation and an ideal arena for semiconductor cavity quantum electrodynamics (cavity QED) experiments as well as coherent control, manipulation, creation and measurement of non-classical states \[2, 11, 15, 16\]. The analysis of nonclassical correlations in semiconductors constitutes a challenging problem, where the physics of interacting electrons must be added to quantum optics and should include properly the effects of noise and dephasing induced by the electron-phonon interaction and the other environment channels \[17\]. The nonlinear optical properties of exciton-cavity system play a key role in driving the quantum correlations and the nonclassical optical phenomena. The crucial role of many-particle Coulomb correlations in semiconductors marks a profound difference from the nonlinear optics of dilute atomic systems, where the optical response is well described by independent transitions between atomic levels, and the nonlinear dynamics is governed only by saturation effects mainly due to the balance of populations between different levels.

The Dynamics Controlled Truncation Scheme (DCTS) provides a (widely adopted) starting point for the microscopic theory of the light-matter interaction effects beyond mean-field \[15\], supplying a consistent and precise way to stop the infinite hierarchy of higher-order correlations which always appears in the microscopic approaches of many-body interacting systems without need to resort to any assumption on the quantum statistics of the quasi-particle arising in due course. By exploiting this scheme, it was possible to express nonlinearities originating from the Coulomb interaction as an instantaneous mean-field exciton-exciton interaction plus a noninstantaneous term where four-particle correlation effects beyond mean-field are contained entirely in a retarded memory function \[2\]. In 1996 the DCTS was extended in order to include in the description the quantization of the electromagnetic field and polariton effects \[18\]. This extension has been applied to the study of quantum optical phenomena in semiconductors and it was exploited to predict polariton entanglement \[13\]. The obtained equations showed that quantum optical correlations (as nonlinear optical effects) arise from both saturation effects (phase-space filling) and Coulomb induced correlations due to four-particle states (including both bound and unbound biexciton states). The dynamical equations included explicitly biexciton states. The structure of those equations didn’t allow the useful separation of Coulomb interaction in terms of a mean-field interaction term plus a noninstantaneous correlation term performed in the semiclassical description.

In this paper we shall set a DCTS microscopic treatment of nonlinear parametric processes in SMCs including the light-field quantization. It represents the starting point for the
microscopic approach to quantum optics experiments in the strong coupling regime. For this purpose we shall exploit a previous technique [2] which, once applied to four-wave mixing in QWs, it allowed to understand a wide range of observed phenomena. Here all the ingredients contributing to the dynamics are introduced and commented. We shall give in great details the manipulations required in order to provide an effective description of the nonlinear parametric contributions beyond mean-field in an exciton-exciton correlation fashion. In particular we derive the coupled equations of motion for the excitonic polarization and the intracavity field. It shows a close analogy to the corresponding equation describing the semiclassical (quantized electron system, classical light field) coherent $\chi^{(3)}$ response in a QW [2], the main difference being that here the (intracavity) light field is regarded not as a driving external source but as a dynamical field [19]. This correspondence is a consequence of the linearization of quantum fluctuations in the nonlinear source term here adopted, namely the standard linearization procedure of quantum correlations adopted for large systems [20]. However the present approach includes the light field quantization and can thus be applied to the description of quantum optical phenomena. Indeed, striking differences between the semiclassical and the full quantum descriptions emerge when considering expectation values of exciton and photon numbers or even higher order correlators, key quantities for the investigation of coherence properties of quantum light [11]. This is the main motivation for the derivation of fully operatorial dynamical equations, within such lowest order nonlinear coherent response, we address in the last section. The results here presented provide a microscopic theoretical starting point for the description of quantum optical effects in interacting electron systems with the great accuracy accomplished for the description of the nonlinear optical response in such many-body systems, see e.g. [2, 15, 19, 21, 22] and references therein. The proper inclusion of the detrimental environmental interaction, an important and compelling issue, is left for a detailed analysis in another paper of ours [23].

In Section II the generality of the coupled system taken into account are exposed, here all the ingredients contributing to the dynamics are introduced and commented. The linear and the lowest nonlinear dynamics is the subject of Sec. III whereas in Sec. IV we shall give in great details the manipulations required in order to provide an effective description of the nonlinear parametric contributions beyond mean-field in an exciton-exciton correlation fashion. In Sec. V the operatorial equations of motion for exciton and intracavity photon operators are derived.
II. THE COUPLED SYSTEM

The system we have in mind is a semiconductor QW grown inside a semiconductor planar Fabry-Perot resonator. In the following we consider a zinc-blende-like semiconductor band structure. The valence band is made from $p$-like ($l = 1$) orbital states which, after spin-orbit coupling, give rise to $j = 3/2$ and $j = 1/2$ decoupled states. In materials like GaAs, the upper valence band is fourfold degenerate ($j = 3/2$), whereas in GaAs-based QWs the valence subbands with $j = 3/2$ are energy splitted into two-fold degenerate heavy valence subbands with $j_z = \pm 3/2$ and light lower energy subbands with $j_z = \pm 1/2$. The conduction band, arising from an $s$-like orbital state ($l=0$), gives rise to $j = 1/2$ twofold states. In the following we will consider for the sake of simplicity only twofold states from the upper valence and lowest conduction subbands. As a consequence electrons in a conduction band as well as holes have an additional spin-like degree of freedom as electrons in free space. When necessary both heavy and light hole valence bands or subbands can be included in the present semiconductor model. Only electron-hole (eh) pairs with total projection of angular momentum $\sigma = \pm 1$ are dipole active in optical interband transitions. In GaAs QWs photons with circular polarizations $\sigma = -(+) \text{ excite electrons with } j^e_z = +1/2 \text{ and holes with } j^h_z = -3/2 \text{ and } j^h_z = 3/2$. We label optically active eh pairs with the same polarization label of light generating them; e.g. $\sigma = +1 \text{ indicates an eh pair with } j^e_z = -1/2$ and $j^h_z = 3/2$.

We start from the usual model for the electronic Hamiltonian of semiconductors [15, 24]. It is obtained from the many-body Hamiltonian of the interacting electron system in a lattice, keeping explicitly only those terms in the Coulomb interaction preserving the number of electrons in a given band, see Appendix B. The system Hamiltonian can be rewritten as

$$\hat{H}_e = \hat{H}_0 + \hat{V}_{\text{Coul}} = \sum_{N\alpha} E_{N\alpha} |N\alpha\rangle\langle N\alpha|,$$

where the eigenstates of $\hat{H}_e$, with energies $E_{N\alpha} = \hbar \omega_{N\alpha}$, have been labelled according to the number $N$ of eh pairs. The state $|N = 0\rangle$ is the electronic ground state, the $N = 1$ subspace is the exciton subspace with the additional collective quantum number $\alpha$ denoting the exciton energy level $n$, the in-plane wave vector $\mathbf{k}$ and the spin index $\sigma$. When needed we will adopt the following notation: $\alpha \equiv (n, k)$ with $k \equiv (\mathbf{k}, \sigma)$. In QWs, light and heavy holes in valence band are split off in energy. Assuming that this splitting is much larger than kinetic energies
of all the involved particles and, as well, much larger than the interaction between them, we shall consider only heavy hole states as occupied. On the contrary to the bulk case, in a QW single particle states experience confinement along the growth direction and subbands appear, anyway in the other two orthogonal directions translational invariance is preserved and the in-plane exciton wave vector remains a good quantum number. Typically, the energy difference between the lowest QW subband level and the first excited one is larger than the Coulomb interaction between particles, and we will consider excitonic states arising from electrons and heavy holes in the lowest subbands.

Eigenstates of the model Hamiltonian with $N=1$ (called excitons) can be created from the ground state by applying the exciton creation operator:

$$|1n\sigma k\rangle = \hat{B}_{n\sigma}^{\dagger} |N = 0\rangle,$$

which can be written in terms of electrons and holes operators as

$$\hat{B}_{n\sigma}^{\dagger} = \sum_k \Phi_{n\sigma}^k \hat{c}_{\sigma,e,k}^{\dagger} \hat{d}_{\sigma,h,k/2}^{\dagger} + \text{h.c.},$$

here $\Phi_{n\sigma}^k$ is the exciton wave function, being $k$ the total wave vector $k = k_e + k_h$, and $k' = \eta_e k_e - \eta_h k_h$ with $\eta_{(e,h)} = m_{(e,h)}/(m_e + m_{(h)})$ ($m_e$ and $m_h$ are the electron and hole effective masses). These exciton eigenstates can be obtained by requiring the general one $eh$ pair states to be eigenstates of $\hat{H}_e$:

$$\hat{H}_e |1n\sigma k\rangle = \hbar \omega_{1n\sigma} |1n\sigma k\rangle,$$

and projecting this secular equation onto the set of product ($eh$) states $|k_e, k_h\rangle = \hat{c}_{k_e}^{\dagger} \hat{d}_{k_h}^{\dagger} |N = 0\rangle$, (see Appendix B for details):

$$\sum_{k_e, k_h} (\langle k_e', k_h' | \hat{H}_e | k_e, k_h\rangle - \hbar \omega_{1n\sigma} \delta_{k_e, k_e'} \delta_{k_h, k_h'}) \langle k_e, k_h | 1n\sigma k\rangle = 0.$$

Thus, having expressed the correlated exciton state as a superposition of uncorrelated product states,

$$|1n\sigma k\rangle = \sum_{k_e, k_h} \langle k_e, k_h | 1n\sigma k\rangle |k_e, k_h\rangle,$$

the scalar products, coefficients of this expansion, represent nothing but the envelope function $\Phi_{n\sigma,k'}^{k}$ of the excitonic aggregate being the solution of the corresponding Schrödinger equation B. It describes the correlated $eh$ relative motion in $k$-space. In order to simplify
a bit the notation, the spin convention in Eq. (3) has been changed by using the same label for the exciton spin quantum number and for the spin projections of the electron and hole states forming the exciton.

The next relevant subspace \((N = 2)\) is the biexciton one, spanning all the states with 2 \(eh\) pairs. It seems worth noting that the above description of \(eh\) complexes arises from the properties of quantum states and, once fixed the system Hamiltonian, no approximations have been introduced insofar. Indeed such a property hold for any \(N\) \(eh\) pairs aggregate and we will give a full account of it in Appendix [B].

The eigenstates of the Hamiltonian \(\hat{H}_c\) of the cavity modes can be written as \(|n, \lambda\rangle\) where \(n\) stands for the total number of photons in the state and \(\lambda = (k_1, \sigma_1; \ldots; k_n, \sigma_n)\) specifies wave vector and polarization \(\sigma\) of each photon. Here we shall neglect the longitudinal-transverse splitting of polaritons [25] originating mainly from the corresponding splitting of cavity modes. It is more relevant at quite high in-plane wave vectors and often it results to be smaller than the polariton linewidths. The present description can be easily extended to include it. We shall treat the cavity field in the quasi-mode approximation, that is to say we shall quantize the field as the mirrors were perfect and subsequently we shall couple the cavity with a statistical reservoir of a continuum of external modes. This coupling is able to provide the cavity losses as well as the feeding of the coherent external impinging pump beam. The cavity mode Hamiltonian, thus, reads

\[
\hat{H}_c = \sum_k \hbar \omega^c_k \hat{a}_k^\dagger \hat{a}_k^{},
\]

with the operator \(\hat{a}_k^\dagger\) which creates a photon state with energy \(\hbar \omega^c_k = \hbar (\omega^2_{\text{exc}} + v^2 |k|^2)^{1/2}\), \(v\) being the velocity of light inside the cavity and \(k = (\sigma, k)\). The coupling between the electron system and the cavity modes is given in the usual rotating wave approximation [18, 26]

\[
\hat{H}_I = - \sum_{nk} V^*_n k \hat{a}_k^\dagger \hat{B}_{nk} + H.c.,
\]

\(V_{n,k}\) is the photon-exciton coupling coefficient enhanced by the presence of the cavity set as \(V_{n,k} = \tilde{V}_\sigma \sqrt{A} \phi^*_{n,\sigma}(x = 0)\), the latter being the real-space exciton envelope function calculated in the origin whereas \(A\) is the in-plane quantization surface, \(\tilde{V}_\sigma\) is proportional to the interband dipole matrix element. Modeling the loss through the cavity mirrors within the quasi-mode picture means we are dealing with an ensemble of external modes, generally
without a particular phase relation among themselves. An input light beam impinging on one of the two cavity mirrors is an external field as well and it must belong to the family of modes of the corresponding side (i.e. left or right). Being coherent, it will be the non zero expectation value of the ensemble. It can be shown [18, 23] that for a coherent input beam, the driving of the cavity modes may be described by the model Hamiltonian [18, 23]

\[ \hat{H}_p = i t_c \sum_k (E_k \hat{a}^\dagger_k - E_k^* \hat{a}_k), \] (9)

where \( E_k \) (\( E_k^* \)) is a \( \mathbb{C} \)-number describing the positive (negative) frequency part of the coherent input light field amplitude.

### III. LINEAR AND NONLINEAR DYNAMICS

The idea is not to use a density matrix approach, but to derive directly expectation values of all the quantities at play. The dynamics is described by “transition” operators (known as generalized Hubbard operators):

\[ \hat{X}_{N,\alpha;M,\beta} = |N, \alpha\rangle \langle M, \beta| \]
\[ \hat{Y}_{n,\lambda;m,\mu} = |n, \lambda\rangle \langle m, \mu|. \] (10)

The fundamental point in the whole analysis is that, thanks to the form of the interaction Hamiltonian \( \hat{H}_I \) and thanks to the quasiparticle conservation the free Hamiltonians possess, we can use the so-called dynamics controlled truncation scheme, stating that we are facing a rather special model where the correlation have their origin only in the action of the electromagnetic field and thus the general theorem due to Axt and Stahl [1] holds. For our purpose we will need its generalization in order to include the quantization of the electromagnetic field [18], it reads:

\[ \langle \hat{X}_{N,\alpha;M,\beta} \hat{Y}_{n,\lambda;m,\mu} \rangle = \sum_{i=0}^{i_0} \langle \hat{X}_{N,\alpha;M,\beta} \hat{Y}_{n,\lambda;m,\mu} \rangle^{(N+M+n+m+2i)} + \mathcal{O}(E^{N+M+n+m+2i_0+2}), \] (11)

i.e. the expectation value of a zero to N-pair transition is at least of order \( N \) in the external electromagnetic field. There are only even powers because of the spatial inversion symmetry which is present. Once a perturbative order in the external coherent fields is
chosen, Eq. (11) limits the expectation values to take into account, thus providing a precise way to truncate the hierarchy of equations of motions.

The exciton and photon operators can be expressed as

\[ \hat{a}_k = \hat{Y}_{0;1k} + \sum_{n \geq 1} \sqrt{n_k + 1} \hat{Y}_{n_k; (n_k + 1)k} \]

\[ \hat{B}_{nk} = \hat{X}_{0;1nk} + \sum_{N \geq 1, \alpha \beta} \langle N\alpha | \hat{B}_{nk} | (N + 1)\beta \rangle \hat{X}_{N\alpha; (N+1)\beta}, \tag{12} \]

where in writing the photon expansion we omitted all the states not belonging to the \( k \)-th mode which add up giving the identity in every Fock sector \[28\].

The equation of motion for the generic quantity of interest \( \hat{X}_{N\alpha; M\beta} \hat{Y}_{n\lambda; m\mu} \) is reported in Appendix A. In the Heisenberg picture we start considering the equation of motion for the photon and exciton operators, once taken the expectation values we exploit theorem (11) retaining only the linear terms. With the help of the generalized Hubbard operators all this procedure may be done by inspection. The linear dynamics for \( \langle \hat{a}_k \rangle^{(1)} = \langle \hat{Y}_{0;1k} \rangle^{(1)} \) and \( \langle \hat{B}_{nk} \rangle^{(1)} = \langle \hat{X}_{0;1nk} \rangle^{(1)} \) reads:

\[ \frac{d}{dt} \langle \hat{a}_k \rangle^{(1)} = -i\bar{\omega}_k \langle \hat{a}_k \rangle^{(1)} + i \sum_n \frac{V_{nk}^*}{\hbar} \langle \hat{B}_{nk} \rangle^{(1)} + t_c \frac{E_k}{\hbar}, \tag{13} \]

\[ \frac{d}{dt} \langle \hat{B}_{nk} \rangle^{(1)} = -i\bar{\omega}_{1nk} \langle \hat{B}_{nk} \rangle^{(1)} + i \frac{V_{nk}}{\hbar} \langle \hat{a}_k \rangle^{(1)}. \tag{14} \]

In these equations \( \bar{\omega}_k = \omega_k - i\gamma_k \), where \( \gamma_k \) is the cavity damping, analogously \( \bar{\omega}_{1nk} = \omega_{1nk} - i\Gamma_x \) and \( \bar{\omega}_{2\beta} = \omega_{2\beta} - i\Gamma_{xx} \). The dynamics up to the third order is a little bit more complex, we shall make extensively use of (A11) (in the following the suffix \( ^{+ (n)} \) stands for “up to” \( n \)-th order terms in the external electromagnetic exciting field). With Eq. (12) the exciton and the photon expectation values can be expanded as follows:

\[ \langle \hat{B}_{nk} \rangle^{+(3)} = \langle \hat{X}_{0;1nk} \rangle^{+(3)} + \sum_{\alpha \beta} \langle 1\alpha | \hat{B}_{nk} | 2\beta \rangle \langle \hat{X}_{1\alpha;2\beta} \rangle^{(3)}, \tag{15} \]

\[ \langle \hat{a}_k \rangle^{+(3)} = \langle \hat{Y}_{0;1k} \rangle^{+(3)} + \sqrt{2} \langle \hat{Y}_{1k;2k} \rangle^{(3)}. \tag{16} \]

With a bit of algebra we obtain

\[ \frac{d}{dt} \langle \hat{a}_k \rangle^{+(3)} = -i\bar{\omega}_k \langle \hat{a}_k \rangle^{+(3)} + i \sum_n \frac{V_{nk}^*}{\hbar} \langle \hat{B}_{nk} \rangle^{+(3)} + t_c \frac{E_k}{\hbar}, \tag{17} \]
\[
\frac{d}{dt} \langle \hat{B}_{nk} \rangle^{(3)} = -i\omega_{1nk} \langle \hat{B}_{nk} \rangle^{(3)} + \frac{V_{nk}}{\hbar} \langle \hat{a}_k \rangle^{(3)} + \\
+ \sum_{\tilde{n}k} \left[ \frac{i}{\hbar} \sum_{n'k',\alpha} V_{n'k'} \langle 1\tilde{n}\tilde{k} | [\hat{B}_{nk}, \hat{B}_{n'k'}^{\dagger}] - \delta_{(n'k');(nk)} | 1\alpha \rangle \langle \hat{X}_{1\tilde{n}\tilde{k};1\alpha} \hat{Y}_{0;1k'} \rangle^{(3)} - \right. \\
\left. -i \sum_{\beta} (\omega_{2\beta} - \omega_{1\tilde{n}} - \omega_{1nk}) \langle 1\tilde{n}\tilde{k} | \hat{B}_{nk} | 2\beta \rangle \langle \hat{X}_{1\tilde{n}\tilde{k};2\beta} \hat{Y}_{0;0} \rangle^{(3)} \right] 
\] 

in analogy with the eqs \[18\] (see also Ref. \[2\]). The resulting equation of motion for the lowest order biexciton amplitude is

\[
\frac{d}{dt} \langle \hat{X}_{0;2\beta} \rangle^{(2)} = -i\omega_{2\beta} \langle \hat{X}_{0;2\beta} \rangle^{(2)} + \\
+ \frac{i}{\hbar} \sum_{n'k',n''k''} V_{n'k'} \langle 2\beta | \hat{B}_{n'k'}^{\dagger} | 1n''k'' \rangle \langle \hat{X}_{0;1n''k''} \hat{Y}_{0;1k'} \rangle^{(2)}. 
\] 

IV. COHERENT RESPONSE

Thanks to the fact we want to treat coherent optical processes it is possible to manipulate further the parametric contributions under two assumptions. We are addressing a coherent optical response, thus we may consider that a coherent pumping mainly generates coherent nonlinear processes, as a consequence the dominant contribution of the biexciton sector on the third-order nonlinear response can be calculated considering the system quantum state as a pure state, which means the nonlinear term is regarded as originating mainly from coherent contributions. Moreover nonclassical correlations are taken into account up to the lowest order. The first assumption results in the factorizations

\[
\langle \hat{X}_{1\tilde{n}\tilde{k};2\beta} \hat{Y}_{0;0} \rangle^{(3)} \approx \langle \hat{X}_{1\tilde{n}k;0} \rangle^{(1)} \langle \hat{X}_{0;2\beta} \rangle^{(2)} \text{ and } \langle \hat{X}_{1\tilde{n}\tilde{k};1\beta} \hat{Y}_{0;1k'} \rangle^{(3)} \approx \langle \hat{X}_{1\tilde{n}k;0} \rangle^{(1)} \langle \hat{X}_{0;1\beta} \hat{Y}_{0;1k'} \rangle^{(2)}. 
\]

The second implies \(\langle \hat{X}_{0;1\beta} \hat{Y}_{0;1k'} \rangle^{(2)} \approx \langle \hat{X}_{0;1\beta} \rangle^{(1)} \langle \hat{Y}_{0;1k'} \rangle^{(1)},\) in the nonlinear source term, namely the standard linearization procedure of quantum correlations adopted for large systems \[20\]. Of course these two approximations can be avoided at the cost of enlarging the set of coupled equations in order to include the equation of motions for the resulting correlation functions. It neglects higher order quantum optical correlation effects between the electron system and the cavity modes leading to a renormalization of the biexciton dynamics with intriguing physical perspectives. However for extended systems, like QWs in planar microcavities, these are effects in most cases of negligible impact, on the contrary in fully confined geometries such as cavity embedded quantum dots they could give significant contributions. In the end, within such a coherent limit, we are able to describe the biexciton
contribution effectively as an exciton-exciton correlation \[2\]. The resulting equations for the coupled exciton an cavity-field expectation values coincide with those obtained within a semiclassical theory (quantized electron-system and classical cavity field). Nevertheless completely different results can be obtained for exciton or photon number expectation values or for higher order correlation function \[13, 26\]. In the next section we will derive operator equations useful for the calculation of such correlation functions. After the two approximations described above (linearization of quantum fluctuations and coherent limit), Eqs (18) becomes

\[
\frac{d}{dt} \langle \hat{B}_{nk} \rangle^{(3)} = -i\bar{\omega}_{1nk} \langle \hat{B}_{nk} \rangle^{(3)} + i \frac{V_{nk}}{\hbar} \langle \hat{a}_k \rangle^{(3)} - i \frac{1}{\hbar} \sum_{\tilde{n}k} \langle \hat{B}_{\tilde{n}k} \rangle^{(1)} R^{(2)}_{nk;\tilde{n}k},
\]

where

\[
R^{(2)}_{nk;\tilde{n}k} = Q^{PSF(2)}_{nk;\tilde{n}k} + Q^{COUL(2)}_{nk;\tilde{n}k},
\]

\[
Q^{PSF(2)}_{nk;\tilde{n}k} = \sum_{n''k''} C^{n''k''}_{nk,\tilde{n}k} \langle \hat{B}_{n''k''} \rangle^{(1)} \langle \hat{a}_{k''} \rangle^{(1)},
\]

\[
Q^{COUL(2)}_{nk;\tilde{n}k} = \sum_{\beta} c^{(1)}_{nk;\tilde{n}k;\beta} \langle \hat{X}_{0;2\beta} \rangle^{(2)},
\]

with

\[
C^{n''k''}_{nk,\tilde{n}k} = V_{n''k''} \langle 1\tilde{n}k | \hat{d}(n''k'')_{nk} - \hat{B}_{nk}^{\dagger} \hat{B}_{n''k''} | 1nk \rangle,
\]

\[
c^{(1)}_{nk;\tilde{n}k;\beta} = \hbar (\omega_{2\beta} - \omega_{1\tilde{n}k} - \omega_{1nk}) \langle 1\tilde{n}k | \hat{B}_{nk} | 2\beta \rangle.
\]

This equation is analogous to the corresponding equation describing the semiclassical (quantized electron system, classical light field) coherent \(\chi^{(3)}\) response in a QW \[2\], the main difference being that here the (intracavity) light field is regarded not as a driving external source but as a dynamical field \[19\]. This close correspondence for the dynamics of expectation values of the exciton operators, is a consequence of the linearization of quantum fluctuations. However the present approach includes the light field quantization and can thus be applied to the description of quantum optical phenomena. By explicit calculation it is easy to see that the first term in Eq. (22) is zero unless all the involved polarization labels \(\sigma\) coincide. In order to manipulate the last term we follow the procedure of Ref. \[2\] which succeeded in reformulating the nonlinear term coming from the Coulomb interaction as an exciton-exciton (X-X) mean-field contribution plus a correlation term driven by a two-exciton correlation function. Even if we are about to perform more or less the same steps
of Ref. [2] we shall provide a detailed account of all the key points of the present derivation. A clear comprehension of these details will be essential for the extension to operatorial dynamical equations of the next section.

In performing this we shall need the two identities:

\[ \sum_{\beta} c_{\beta, n_k}^{(1)} \langle \tilde{x}_0, \beta \rangle^{(2)} = \hbar \sum_{\beta} \langle 1 n_k \mid \hat{B}_{n_k} \rangle \left( \frac{\hat{H}_e}{\hbar} - \omega_{n_k} \right) \langle 2 \beta \rangle \cdot \]

\[ \frac{1}{2} \sum_{n_k', n_{k''}} \left( \langle \hat{B}_{n_k'} \rangle^{(1)} \langle \hat{B}_{n_{k''}} \rangle^{(1)} e^{-i n_{k''}} \right) = \frac{i}{\hbar} \sum_{n_k', n_{k''}} \langle \hat{B}_{n_k'} \rangle^{(1)} \langle \hat{B}_{n_{k''}} \rangle^{(1)} e^{-i n_{k''}} \cdot \]

where \( \Omega = \omega_{n_{k'}} + \omega_{n_{k''}} - 2i \Gamma_x \). Employing the forma solution of the biexciton amplitude Eq. (19) we have:

\[ \sum_{\beta} c_{\beta, n_k}^{(1)} \langle \hat{B}_{n_k} \rangle^{(1)} \langle \hat{B}_{n_{k''}} \rangle^{(1)} e^{-i \Omega(t-t')} = \left( \frac{1}{2} \right) \sum_{n_k', n_{k''}} \langle \hat{B}_{n_k'} \rangle^{(1)} \langle \hat{B}_{n_{k''}} \rangle^{(1)} e^{-i \Omega(t-t')} \cdot \]

We observe that the matrix elements entering the nonlinear source terms are largely independent on the wave vectors for the range of wave vectors of interest in the optical response. Neglecting such dependence we can thus exploit the identity (26), obtaining

\[ \sum_{\beta} c_{\beta, n_k}^{(1)} \langle \hat{B}_{n_k} \rangle^{(1)} \langle \hat{B}_{n_{k''}} \rangle^{(1)} e^{-i \Omega(t-t')} = \left( \frac{1}{2} \right) \sum_{n_k', n_{k''}} \langle \hat{B}_{n_k'} \rangle^{(1)} \langle \hat{B}_{n_{k''}} \rangle^{(1)} e^{-i \Omega(t-t')} \cdot \]
where in the last lines we have resummed all the biexciton subspace by virtue of its completeness. By performing an integration by part, Eq. (28) can be rewritten as

\[
\begin{align*}
&= \frac{1}{2} \hbar \sum_{n'k',n''k''} \left\{ e^{i(\omega_{1n'k'} + \omega_{1n'k''} - 2i\Gamma_x + i\Gamma_{xx})(t-t')} \langle \tilde{1} \tilde{n} \tilde{k} | \hat{B}_{nk} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk} \right) e^{-i\frac{\hat{H}_c}{\hbar}(t-t')} \hat{B}_{n'k'}^\dagger | 1n''k'' \rangle \\
&\quad \left( \hat{B}_{n'k'} \right)^{(1)} (t') \langle \hat{B}_{n''k''} \rangle^{(1)} (t') e^{-i\Omega(t-t')} \right\} \bigg|_{-\infty}^t - \\
&\quad - \int_{-\infty}^t dt' \langle \hat{B}_{n'k'} \rangle^{(1)} (t') \langle \hat{B}_{n''k''} \rangle^{(1)} (t') e^{-i\Omega(t-t')} \frac{d}{dt'} \left\{ e^{i(\omega_{1n'k'} + \omega_{1n'k''} - 2i\Gamma_x + i\Gamma_{xx})(t-t')} \langle \tilde{1} \tilde{n} \tilde{k} | \hat{B}_{nk} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk} \right) e^{-i\frac{\hat{H}_c}{\hbar}(t-t')} \hat{B}_{n'k'}^\dagger | 1n''k'' \rangle \right\}
\end{align*}
\]

The first and the second term can be expressed in terms of a double commutator structure:

\[
\langle \tilde{1} \tilde{n} \tilde{k} | \hat{B}_{nk} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk} \right) \rangle = \langle 0 | [\hat{B}_{\tilde{n}\tilde{k}}, \hat{H}_c] = \langle 0 | \hat{D}_{\tilde{n}\tilde{k},nk},
\]

where a force operator \( \hat{D} \) is defined and

\[
\frac{d}{dt'} \left\{ e^{i(\omega_{1n'k'} + \omega_{1n'k''} - 2i\Gamma_x + i\Gamma_{xx})(t-t')} \langle \tilde{1} \tilde{n} \tilde{k} | \hat{B}_{nk} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk} \right) e^{-i\frac{\hat{H}_c}{\hbar}(t-t')} \hat{B}_{n'k'}^\dagger | 1n''k'' \rangle \right\} =
\]

\[
\begin{align*}
&= \frac{d}{dt'} \left\{ \langle 0 | \hat{D}_{\tilde{n}\tilde{k},nk} e^{-i\frac{\hat{H}_c}{\hbar}(t-t')} \hat{B}_{n'k'}^\dagger \hat{B}_{n''k''}^\dagger | 0 \rangle e^{i(\omega_{1n'k'} + \omega_{1n'k''} - 2i\Gamma_x + i\Gamma_{xx})(t-t')} \right\} = \\
&= \langle 0 | \hat{D}_{\tilde{n}\tilde{k},nk} e^{-i\frac{\hat{H}_c}{\hbar}(t-t')} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1n'k'} - \omega_{1n'k''} - i(\Gamma_{xx} - 2\Gamma_x) \right) \hat{B}_{n'k'}^\dagger \hat{B}_{n''k''}^\dagger | 0 \rangle e^{i(\omega_{1n'k'} + \omega_{1n'k''} - 2i\Gamma_x + i\Gamma_{xx})(t-t')} = \\
&= e^{i(\omega_{1n'k'} + \omega_{1n'k''} - 2i\Gamma_x + i\Gamma_{xx})(t-t')} i F^n_{n',n''k',n''k'}^{n',n''k'}(t-t') + (\Gamma_{xx} - 2\Gamma_x) e^{i(\omega_{1n'k'} + \omega_{1n'k''} - 2i\Gamma_x + i\Gamma_{xx})(t-t')} \langle 0 | \hat{D}_{\tilde{n}\tilde{k},nk} (t-t') \hat{B}_{n'k'}^\dagger \hat{B}_{n''k''}^\dagger | 0 \rangle,
\end{align*}
\]
where the memory kernel reads

\[ F_{\tilde{n}k, n k}^{n''k'', n'k'}(t - t') = \langle 0 | \hat{D}_{\tilde{n}k, nk}(t - t') \hat{D}_{n''k'', n'k'}^{\dagger} | 0 \rangle. \]  

(33)

The usual time dependence in the Heisenberg picture is given by \( \hat{D}(\tau) = e^{i(\hat{H}_c/\hbar)\tau} \hat{D} e^{-i(\hat{H}_c/\hbar)\tau}. \) Altogether, the nonlinear term originating from Coulomb interaction can be written as

\[
Q_{nk; \tilde{n}k}^{\text{COUL}(2)} = \sum_{\beta} c_{nk; \tilde{n}k; \beta}^{(1)} \langle \hat{X}_{0;2\beta} \rangle^{(2)} = \\
\frac{1}{2} \hbar \sum_{n'k'; n''k''} \left\{ \langle 0 | \hat{D}_{\tilde{n}k, nk} \hat{B}_{n'k'}^{\dagger} \hat{B}_{n''k''}^{\dagger} | 0 \rangle \langle \hat{B}_{n'k'} \rangle^{(1)}(t) \langle \hat{B}_{n''k''} \rangle^{(1)}(t) - i \int_{-\infty}^{t} dt' F_{\tilde{n}k, nk}^{n''k'', n'k'}(t - t') \langle \hat{B}_{n'k'} \rangle^{(1)}(t') \langle \hat{B}_{n''k''} \rangle^{(1)}(t') e^{-\Gamma_{xx}(t-t')} \right\} - \hbar \left( \Gamma_{xx} - 2\Gamma_x \right) \sum_{n'k'; n''k''} \int_{-\infty}^{t} dt' \langle 0 | \hat{D}_{\tilde{n}k, nk}(t - t') \hat{B}_{n'k'}^{\dagger} \hat{B}_{n''k''}^{\dagger} | 0 \rangle \langle \hat{B}_{n'k'} \rangle^{(1)}(t') \langle \hat{B}_{n''k''} \rangle^{(1)}(t') .
\]

(34)

For later purpose we are interested in the optical response dominated by the 1S exciton sector, with \( \Gamma_{xx} \simeq 2\Gamma_x \) in the cases of counter- and co-circularly polarized waves. Specifying to this case the Coulomb-induced term with Eq. (34) becomes

\[
\frac{d}{dt} \langle \hat{B}_{\pm k} \rangle^{+^{(3)}}_{\text{COUL}} = -i\bar{\omega}_k \langle \hat{B}_{\pm k} \rangle^{+^{(3)}}_{\text{COUL}} - i \hbar \sum_{\tilde{\sigma}k} \langle \hat{B}_{\tilde{\sigma}k} \rangle^{*^{(1)}}(1) Q_{\pm; k; \tilde{\sigma}k}^{\text{COUL}(2)} = \\
- i\bar{\omega}_k \langle \hat{B}_{\pm k} \rangle^{+^{(3)}}_{\text{COUL}} - i \hbar \sum_{k; \pm; k; k; k'} \delta_{k+k'-k''} V_{xx} \langle \hat{B}_{\pm k} \rangle^{*^{(1)}}(t) \langle \hat{B}_{\pm k} \rangle^{(1)}(t) \langle \hat{B}_{\pm k} \rangle^{(1)}(t) + \\
- \frac{1}{\hbar} \sum_{k; k; k; k; k''} \delta_{k+k'-k''} \delta_{\pm; \pm; \sigma; \sigma'} \langle \hat{B}_{\tilde{\sigma}k} \rangle^{*^{(1)}}(t) \int_{-\infty}^{t} dt' F_{\sigma; \sigma''}(t - t') \langle \hat{B}_{\sigma' k'} \rangle^{(1)}(t') \langle \hat{B}_{\sigma'' k''} \rangle^{(1)}(t') e^{-\Gamma_{xx}(t-t')} ,
\]

(35)

where, in order to lighten the notation, we dropped the two spin indexes \( \sigma \) and \( \tilde{\sigma} \) in the four-particle kernel function \( F \) defined in Eq. (33) for they are already univocally determined once chosen the others (i.e. \( \sigma' \) and \( \sigma'' \)) as soon as their selection rule \( \langle \delta_{\sigma+\tilde{\sigma}; \sigma'+\sigma''} \rangle \) is applied. Moreover, the \( \hbar/2 \) has been reabsorbed in the Coulomb nonlinear coefficients \( V_{xx} \) and \( F_{\sigma; \sigma''}(t - t') \). A detail microscopic account for the mean-field \( V_{xx} \), for the \( F \)'s and their selection rules are considered in [29, 30]. For the range of \( k \)-space of interest, i.e. \( |k| \ll \frac{\alpha}{a_x} \) (much lower than the inverse of the exciton Bohr radius) they are largely independent on the center of mass wave vectors. While \( V_{xx} \) and \( F^{\pm; \pm}(t - t') \) (i.e. co-circularly polarized waves)
conserve the polarizations, $F^\pm\mp(t - t')$ and $F^\mp\pm(t - t')$ (counter-circular polarization) give rise to a mixing between the two circularly polarizations. The physical origin of the three terms in Eq. (34) can be easily understood: the first is the Hartee-Fock or mean-field term representing the first order treatment in the Coulomb interaction between excitons, the second term is a pure biexciton (four-particle correlations) contribution. This coherent memory may be thought as a non-Markovian process involving the two-particle (excitons) states interacting with a bath of four-particle correlations [2]. Equation (34) even if formally similar to that of Ref. [2], represents its extension including polaritonic effects due to the presence of the cavity. It has been possible thanks to the inclusion of the dynamics of the cavity modes whereas in Ref. [2], the electromagnetic field entered as a parameter only. Former analogous extensions have been obtained within a semiclassical model [19, 29, 30]. The strong exciton-photon coupling does not modify the memory kernel because four-particle correlations do not couple directly to cavity photons. As pointed out clearly in Ref. [19], cavity effects alter the phase dynamics of two-particle states during collisions, indeed, the phase of two-particle states in SMCs oscillates with a frequency which is modified respect to that of excitons in bare QWs, thus producing a modification of the integral in Eq. (34). In this way the exciton-photon coupling $V_{nk}$ affects the exciton-exciton collisions that govern the polariton amplification process. Ref. [19] considers the first (mean-field) and the second (four-particle correlation) terms in the particular case of cocircularly polarized waves, calling them without indexes as $V_{xx}$ and $F(t)$ respectively. In Fig. 1 they show $\mathcal{F}(\omega)$, the Fourier transform of $F(t)$ plus the mean-field term $V_{xx}$, 

$$\mathcal{F}(\omega) = V_{xx} - i \int_{-\infty}^{\infty} dt F(t)e^{i\omega t}.$$ 

(36)

Its imaginary part is responsible for the frequency dependent excitation induced dephasing, it reflects the density of the states of two-exciton pair coherences. Towards the negative detuning region the dispersive part $\text{Re}(\mathcal{F})$ increases whereas the absorptive part $\text{Im}(\mathcal{F})$ goes to zero. The former comprises the mean-field contribution effectively reduced by the four-particle contribution. Indeed, the figure shows the case with a binding energy of 13.5 meV, it gives $V_{xx}n_{sat} \simeq 11.39$ meV which clearly is an upper bound for $\text{Re}(\mathcal{F})$ for negative detuning. The contribution carried by $F(t)$ determines an effective reduction of the mean-field interaction (through its imaginary part which adds up to $V_{xx}$) and an excitation induced dephasing. It has been shown [19] that both effects depends on the sum of the energies of
the scattered polariton pairs. The third term in Eq. \(34\) can be thought as a reminder of the mismatch between the picture of a biexciton as a composite pair of exciton. In the following we will set \(\Gamma_{xx} \approx 2\Gamma_x\).

The other nonlinear source term in Eq. \(21\) depends directly on the exciton wave function and reads

\[
\sum_{\tilde{n}\tilde{k}} (\tilde{B}_{\tilde{n}\tilde{k}})^{(1)} \sum_{n'k',n''k''} C_{\tilde{n}\tilde{k},nk}^{n'k',n''k''} \langle \tilde{\alpha}_{k'} \rangle^{(1)} \langle \tilde{B}_{n''k''} \rangle^{(1)} .
\]

(37)

It represents a phase-space filling (PSF) contribution, due to the Pauli blocking of electrons. It can be developed as follows,

\[
C_{\tilde{n}\tilde{k},nk}^{n'k',n''k''} = V_{n'k'} \langle \tilde{\alpha}_{n'k'} | \delta_{(n'k');(nk)} - [\tilde{B}_{n\sigma k}, \tilde{B}_{n'\sigma' k'}^\dagger] | 1n''\sigma''k'' \rangle =
\]

\[
= V_{n'k'} \delta_{\sigma,\sigma'} \left\{ \sum_q \Phi_{n\sigma q}^* \Phi_{n'\sigma' q}^{(k+k')} \langle \tilde{\alpha}_{n'\sigma'q} | \delta_{\sigma',q,q+k} C_{\sigma,q+k}^* C_{\sigma,q+k} | 1n''\sigma''k'' \rangle \right. +
\]

\[
\sum_q \Phi_{n\sigma q}^* \Phi_{n'\sigma' q}^{(k+k')} \langle \tilde{\alpha}_{n'\sigma'q} | \delta_{\sigma',q,q+k} C_{\sigma,q+k}^* C_{\sigma,q+k} | 1n''\sigma''k'' \rangle \right\} =
\]

\[
= V_{n'k'} \delta_{\sigma,\sigma'} \delta_{\tilde{n}+k,k+k''} \left\{ \sum_q \Phi_{n\sigma q}^* \Phi_{n'\sigma' q}^{(k+k')} \Phi_{n''\sigma'' q}^{(k+k'')} +
\right.
\]

\[
\sum_q \Phi_{n\sigma q}^* \Phi_{n'\sigma' q}^{(k+k')} \Phi_{n''\sigma'' q}^{(k+k'')} \right\} ,
\]

(38)

the explicit expressions of the \(q\)'s are given in \[31\].

Thus, the nonlinear dynamics of Eq. \(20\) driven by \(\tilde{H}_I\) can be written

\[
\frac{d}{dt} \langle \tilde{B}_{n\sigma k} \rangle^{(1)} |_{\tilde{H}_I} = +i \frac{V_{n\sigma k}}{\hbar} \langle \tilde{\alpha}_{\sigma k} \rangle^{(1)} - i \frac{\hbar}{2} \sum_{n'n''k'k''} \delta_{k+k',k''} \langle \tilde{B}_{n'\sigma k'} \rangle^{(1)} \langle \tilde{B}_{n''\sigma k''} \rangle^{(1)} \langle \tilde{\alpha}_{\sigma k} \rangle^{(1)} \sum_q \Phi_{n\sigma q}^* \Phi_{n'\sigma q}^{(k+k')} \Phi_{n''\sigma q}^{(k+k'')} +
\]

\[
\sum_q \Phi_{n\sigma q}^* \Phi_{n'\sigma q}^{(k+k')} \Phi_{n''\sigma q}^{(k+k'')} \right\} .
\]

(39)

We are interested in studying polaritonic effects in SMCs where the optical response involves mainly excitons belonging to the 1S band with wave vectors close to normal incidence, i.e. \(|k| \ll \frac{c}{\alpha_x}\) (much lower than the inverse of the exciton Bohr radius). In this case the exciton relative wave functions are independent on spins as well as on the center of mass wave vector. They are such that \(\sum_{q=-\infty}^{\infty} |\Phi_q|^2 = 1\), i.e. \(\Phi_q = \frac{1}{\sqrt{A}} \frac{\sqrt{2\pi} a_x}{(1+(\alpha_x|q|)^2)^{1/4}}\), \(a_x\) is the exciton Bohr radius. From now on whenever no excitonic level is specified the 1S label is understood.
It yields
\[
\frac{d}{dt} \langle \hat{B}_{\sigma k} \rangle^{(3)} \bigg|_{\hat{H}_I} = +i \frac{V_{\sigma k}}{\hbar} \langle \hat{a}_{\sigma k} \rangle^{(3)} - \frac{i}{\hbar} \sum_{k'k''k} \delta_{k+k'k''} \langle \hat{B}_{\sigma k} \rangle^{(1)} \langle \hat{a}_{\sigma k'} \rangle^{(1)} \langle \hat{a}_{\sigma k''} \rangle^{(1)} 2\tilde{V}^*O_{PSF},
\]
where the overlap \(O_{PSF}\) has been calculated in the case of zero center of mass wave vector, namely
\[
O_{PSF} = \sum_q \Phi_q^* \Phi_q^* \Phi_q.
\]
In SMCs a measured parameter is the so-called vacuum Rabi splitting \(V_{\sigma k}\) of the 1S excitonic resonance, for the range of \(k\)-space of interest essentially constant. Defining \(V = V_{\sigma} = \tilde{V}_{\sigma} \sqrt{A\phi^*(0)}\)
\[
\tilde{V}_{\sigma}^*O_{PSF} = \frac{V}{\sqrt{A\phi^*(0)}} \frac{8\pi a^2_x}{V} = \frac{1}{2} V_{\text{sat}},
\]
where we have set \(n_{\text{sat}} = (7/16)(A/\pi a^2_x)\), called saturation density.

In terms of the two circular polarizations the dynamics induced by \(\hat{H}_I\) finally reads
\[
\frac{d}{dt} \langle \hat{B}_{\pm k} \rangle^{(3)} \bigg|_{\hat{H}_I} = -i \tilde{\omega}_k \langle \hat{B}_{\pm k} \rangle^{(3)} + i \frac{V}{\hbar} \langle \hat{a}_{\pm k} \rangle^{(3)} - \frac{i}{\hbar} \sum_{\sigma\pm} \langle \hat{B}_{\sigma k} \rangle^{(1)} Q_{\pm k;\sigma k}^{PSF(2)}(1).
\]
where
\[
\sum_{k} \langle \hat{B}_{\pm k} \rangle^{(1)} Q_{\pm k;\sigma k}^{PSF(2)} = \frac{V}{n_{\text{sat}}} \sum_{k'k''k} \delta_{k+k'k''} \langle \hat{B}_{\pm k} \rangle^{(1)} \langle \hat{a}_{\pm k'} \rangle^{(1)} \langle \hat{a}_{\pm k''} \rangle^{(1)}.
\]
The same lines of argument can be followed for computing the Coulomb-induced interactions \(Q_{\text{Coul}}^{(2)}\).

We are lead to introduce the saturation density for two main reasons. The most obvious is our interest to refer this work to the literature where \(n_{\text{sat}}\) is extensively used. The other most interesting reason is that we can directly compute this quantity. Indeed, the equation of motion for the exciton operator reads
\[
\frac{d}{dt} \langle \hat{B}_{\pm k} \rangle^{(3)} = -i \omega_k \langle \hat{B}_{\pm k} \rangle^{(3)} + i \frac{V}{\hbar} \langle \hat{a}_{\pm k} \rangle^{(3)} - \frac{i}{\hbar} \sum_{\sigma\pm} \langle \hat{B}_{\sigma k} \rangle^{(1)} Q_{\pm k;\sigma k}^{Coul(2)(1)}
\]

Leaving apart the discrepancy between the order in the DCTS we can compute the so-called oscillator strength (OS), defined as what multiplies the photon expectation values \(\langle \hat{a}_{\pm k=0} \rangle\),
\[
OS = i \frac{V}{\hbar} \left( 1 - \frac{2}{\sqrt{A\phi^*(0)}} \frac{O_{PSF}}{\langle \hat{B}_{\pm 0} \rangle^{(1)} \langle \hat{B}_{\pm 0} \rangle^{(1)}} \right).
\]
The saturation density may be defined as the exciton density that makes the oscillator strength to be zero. We obtain

\[
    n_{\text{sat}} = \left( \frac{2}{\sqrt{A\phi^*(0)}} O_{\text{PSF}}^{\text{PSF}} \right)^{-1} = \frac{A}{\pi a_x^2} \frac{7}{16}.
\]  

(45)

Eventually, the lowest order \( \chi^{(3)} \) nonlinear optical response in SMCs are described by the following set of coupled equations:

\[
    \frac{d}{dt} \langle \hat{a}_{\pm k} \rangle^{\text{(3)}} = -i \omega_k \langle \hat{a}_{\pm k} \rangle^{\text{(3)}} + i \frac{V}{\hbar} \langle \hat{B}_{\pm k} \rangle^{\text{(3)}} + t_c \frac{E_{\pm k}}{\hbar},
\]

(46)

\[
    \frac{d}{dt} \langle \hat{B}_{\pm k} \rangle^{\text{(3)}} = -i \tilde{\omega}_k \langle \hat{B}_{\pm k} \rangle^{\text{(3)}} + i \frac{V}{\hbar} \langle \hat{a}_{\pm k} \rangle^{\text{(3)}} - i \frac{1}{\hbar} \sum_{\tilde{\sigma}\tilde{k}} \langle \hat{B}_{\tilde{\sigma}\tilde{k}} \rangle^{\text{(1)}} R_{\pm k,\tilde{\sigma}\tilde{k}},
\]

(47)

with \( \sum_{\tilde{\sigma}\tilde{k}} \langle \hat{B}_{\tilde{\sigma}\tilde{k}} \rangle^{\text{(1)}} R_{\pm k,\tilde{\sigma}\tilde{k}} = \sum_{\tilde{\sigma}\tilde{k}} \langle \hat{B}_{\tilde{\sigma}\tilde{k}} \rangle^{\text{(1)}} Q_{\text{COUL}}^{\text{(2)}} + \sum_{\tilde{k}} \langle \hat{B}_{\pm \tilde{k}} \rangle^{\text{(1)}} Q_{\text{PSF}}^{\text{(2)}} \), with the first of the two addenda originating from Coulomb interaction, Eq. (35), whereas the second represents the phase-space filling contribution written in Eqs. (43). Starting from here, in the strong coupling case, it might be useful to transform the description into a polariton basis. The proper inclusion of dephasing/relaxation and the application of these equations to parametric processes, in the strong coupling regime, is described in another paper of ours [23].

Equations (46) and (47) is exact to the third order in the exciting field. While a systematic treatment of higher-order optical nonlinearities would require an extension of the equations of motions (see e.g. Appendix), a restricted class of higher-order effects can be obtained from solving equations (46) and (47) self-consistently up to arbitrary order as it is usually employed in standard nonlinear optics. This can be simply accomplished by replacing, in the nonlinear sources, the linear excitonic polarization and light fields with the total fields \( [2, 19, 22] \). Multiple-scattering processes are expected to be very effective in cavity-embedded QWs due to multiple reflections at the Bragg mirrors.

V. PARAMETRIC PHOTOLUMINESCENCE: TOWARDS SEMICONDUCTOR QUANTUM OPTICS

Entanglement is one of the key features of quantum information and communication technology [4]. Parametric down-conversion is the most frequently used method to generate
highly entangled pairs of photons for quantum-optics applications, such as quantum cryptography and quantum teleportation. This $\chi^{(3)}$ optical nonlinear process consists of the scattering of two polaritons generated by a coherent pump beam into two final polariton modes. The total energy and momentum of the final pairs equal that of pump polariton pairs. The scattering can be spontaneous (parametric emission) or stimulated by a probe beam resonantly exciting one of the two final polariton modes. In 2005 an experiment probing quantum correlations of (parametrically emitted) cavity polaritons by exploiting quantum complementarity has been proposed and realized [11]. The most common set-up for parametric emission is the one where a single coherent pump feed resonantly excites the structure at a given energy and wave vector, $k_p$. Within the DCTS we shall employ Eqs. (17), (18) and Eq. (19) in operatorial form, provided all the equations to become fully significant as soon as the expectation value quantities we shall work out would lie within the consistent perturbative DCTS order we set from the beginning [26]. In order to be more specific we shall derive explicitly the case of input light beams activating only the $1S$ exciton sector with all the same circularly (e.g. $\sigma^+$) polarization, thus excluding the coherent excitation of bound two-pair coherences (biexciton) mainly responsible for polarization-mixing [2]. Equations involving polariton pairs with opposite polarization can be derived in complete analogy following the same steps. Starting from the Heisenberg equations for the exciton and photon operators and keeping only terms providing lowest order nonlinear response (in the input light field) we obtain,

$$\frac{d}{dt} \hat{a}_k = -i\omega_k \hat{a}_k + i \frac{V_k^*}{\hbar} \hat{B}_k + t_c \frac{E_k}{\hbar}, \quad (48)$$

$$\frac{d}{dt} \hat{B}_k = -i\omega_k \hat{B}_k + i \frac{V_k}{\hbar} \hat{a}_k +$$

$$+ \frac{i}{\hbar} \sum_{k,k',\alpha} V_{k'}(1\tilde{k} \mid [\hat{B}_{k'} \hat{B}^\dagger_k] - \delta_{(k'),(k)} \mid 1\alpha) \hat{X}_{1\tilde{k},0} \hat{X}_{0,1\alpha} \hat{Y}_{0,1\beta} -$$

$$- \frac{i}{\hbar} \sum_{k\beta} (\omega_{2\beta} - \omega_{1\tilde{k}} - \omega_{1k}) \langle 1\tilde{k} \mid \hat{B}_k \mid 2\beta \rangle \hat{X}_{1\tilde{k},0} \hat{X}_{0,2\beta}. \quad (49)$$

In the following we will assume that the pump polaritons driven by a quite strong coherent input field consists of a classical ($C$-number) field. This approximation is in close resemblance to the two approximations performed in the previous section (linearization of fluctuations and coherent nonlinear processes). We shall show that under this approximation, we may
perform the same manipulations ending up to a set of coupled equations analogous to Eqs. (46) and (47). In addition, having a precise set-up chosen, we will be able to specialize our equations and give an explicit account of the parametric contributions as well as the shifts the lowest order nonlinear dynamics provides. We shall retain only those terms containing the semiclassical pump amplitude at \( k_p \) twice, thus focusing on the “direct” pump-induced nonlinear parametric scattering processes. It reads

\[
\frac{d}{dt} \hat{B}_{\pm k} = -\omega_k \hat{B}_{\pm k} + i \frac{V}{\hbar} \hat{a}_{\pm k} - \frac{i}{\hbar} \frac{V}{n_{\text{sat}}} \sum_{k,k',k''} \delta_{k+k',k''} \hat{X}_{1 \pm k,0} \hat{X}_{0,1 \pm k'} (\delta_{k'',k_p} \delta_{k',k_p} + \delta_{k,k_p} \delta_{k',k''}) - \frac{i}{\hbar} \sum_{\sigma \beta} (\omega_{2k,\sigma} - \omega_{1k}) \langle 1 \sigma \k \mid \hat{B}_{\pm k} \mid 2 \sigma \beta k_p \rangle \hat{X}_{1 \sigma \k,0} \hat{X}_{0,2 \sigma \beta k_p} \delta_{k,2k_p} + \delta_{k,k_p} \delta_{k',k+k_p}),
\]

where we have already manipulated the phase-space filling matrix element. Here in brackets the first addendum of each line would be responsible for the parametric contribution, whereas the others will give the shifts. It is understood, from now on, that the pump-driven terms (e.g. the \( X \) and \( Y \) at \( k_p \)) are \( \mathbb{C} \)-numbers coherent amplitudes like the semiclassical electromagnetic pump field, we will make such distinction in marking with a “hat” the operators only.

We need some care in manipulating the Coulomb-induced terms, the last line. Written explicitly it is

\[
\frac{d}{dt} \hat{B}_{\pm k} \bigg|_{\text{Coul}} = -\frac{i}{\hbar} \sum_{\sigma \beta} (\omega_{2k,\sigma} - \omega_{1k}) \langle 1 \sigma \k \mid \hat{B}_{\pm k} \mid 2 \sigma \beta k_p \rangle \hat{X}_{1 \sigma \k,0} \hat{X}_{0,2 \sigma \beta k_p} + \\
-\frac{i}{\hbar} \sum_{\sigma \beta} (\omega_{2k,\sigma} - \omega_{1k}) \langle 1 \sigma \k \mid \hat{B}_{\pm k} \mid 2 \sigma \beta k_p \rangle X_{1 \sigma \k,0} X_{0,2 \sigma \beta k_p,0} X_{0,2 \sigma \beta k_p + k+k_p}.
\]  

As for the term containing \( X_{0,2k_p} \), we are facing a \( \mathbb{C} \)-number which gives no problem in performing the very same procedure of the previous chapter. As for the other we would
exploit the formal biexciton solution

\[
\dot{X}_{0:2(k+k_p)}(t) = \int_{-\infty}^{t} dt' e^{-i\omega_{2(k+k_p)}(t-t')} \frac{i}{\hbar} \left( V_{k_p} \langle 2(k+k_p) | \hat{B}_{k_p}^\dagger | 1k \rangle \dot{X}_{0,1k} Y_{0,1k_p} + \right.
\]

\[
V_k \langle 2(k+k_p) | \hat{B}_{k_p}^\dagger | 1k_p \rangle X_{0,1k} \dot{Y}_{0,1k} \right), \tag{52}
\]

where, for the sake of consistence, we are neglecting \( \dot{X}_{0:2(k+k_p)}(-\infty) \) because the biexciton, within the present approximations, is always generated by an operator at \( k \) times a classical amplitude at \( k_p \) which is always zero before the electromagnetic impulse arrived. Moreover, an analogous identity such that of Eq. \((25)\) is valid in the present context, namely

\[
\frac{d}{dt} \left( \dot{X}_{0,1k} X_{0,1k_p} e^{-i(\omega_{1k}+\omega_{1k_p})(t-t')} \right) = \left( \frac{i}{\hbar} V_k \dot{Y}_{0,1k} X_{0,1k_p} + i \frac{V_{k_p}}{\hbar} Y_{0,1k_p} \dot{X}_{0,1k} \right) e^{-i(\omega_{1k}+\omega_{1k_p})(t-t')} \tag{53}
\]

With these tools at hand we are able to perform step by step the manipulations of the previous section for all the quantities at play. The final result reads

\[
\frac{d}{dt} \hat{B}_{\pm k} = -\omega_k \hat{B}_{\pm k} + i \frac{V}{\hbar} \hat{a}_{\pm k} -
\]

\[
- i \frac{V}{\hbar n_{sat}} \left( \dot{X}_{1:1k_{i,0}X_0,1k_p} Y_{0,1k_p} + X_{1:1k_{i,0}X_0,1k_p} \dot{Y}_{0,1k} + X_{1:1k_{i,0}\dot{X}_{0,1k}} Y_{0,1k_p} \right) -
\]

\[
- \frac{i}{\hbar} \dot{X}_{1:1k_{i,0}}(t) \left\{ V_{xx} X_{0,1k_p} (t) X_{0,1k_p} (t) - i \int_{-\infty}^{t} dt' F_{\pm \pm}(t-t') X_{0,1k_p} (t') X_{0,1k_p} (t') \right\} -
\]

\[
-2 \frac{i}{\hbar} X_{1:1k_{i,0}}(t) \left\{ V_{xx} \dot{X}_{0,1k} (t) X_{0,1k_p} (t) - i \int_{-\infty}^{t} dt' F_{\pm \pm}(t-t') X_{0,1k} (t') X_{0,1k_p} (t') \right\} \tag{54}
\]

where \( \mathbf{k_i} = 2\mathbf{k_p} - \mathbf{k} \), and again \( V_{xx} \) and \( F_{\pm \pm}(t-t') \) have reabsorbed the 1/2 originating from Eq. \((53)\). In the specific case under analysis we are considering co-circularly polarized waves and the mean field term, \( V_{xx} \) as well as the the kernel function \( F(t) \) can be found in Refs. \([29, 30]\).

Eventually, the lowest order \( (\chi^{(3)}) \) nonlinear optical response in SMCs is given by the following set of coupled equations where, in the same spirit of the final remark in the previous section, we account for multiple scattering simply by replacing the linear excitonic polarization and light fields with the total fields:

\[
\frac{d}{dt} \hat{a}_{\pm k} = -i\omega_k \hat{a}_{\pm k} + i \frac{V}{\hbar} \hat{B}_{\pm k} + t_c \frac{E_{\pm k}}{\hbar}
\]

\[
\frac{d}{dt} \hat{B}_{\pm k} = -i\omega_k \hat{B}_{\pm k} + \hat{s}_{\pm k} + i \frac{V}{\hbar} \hat{a}_{\pm k} - \frac{i}{\hbar} \hat{P}^{NL}_{\pm k} \tag{55}
\]
where $R_{\pm k}^{NL} = (R_{\pm k}^{sat} + R_{\pm k}^{xx})$

\[
R_{\pm k}^{sat} = \frac{V}{n_{sat}} B_{\pm k_p} a_{\pm k} \hat{B}_{\pm k}^+,
\]

\[
R_{\pm k}^{xx} = \hat{B}_{\pm k}^+(t) \left( V_{xx} B_{\pm k_p}(t) B_{\pm k_p}(t) - i \int_{-\infty}^{t} dt' F_{\pm\pm}(t - t') B_{\pm k_p}(t') B_{\pm k_p}(t') \right). \tag{56}
\]

The pump induced renormalization of the exciton dispersion gives a frequency shift

\[
\hat{s}_{\pm k} = -i \left( \frac{V}{n_{sat}} (B_{\pm k_p}^* a_{\pm k} \hat{B}_{\pm k} + B_{\pm k_p}^* B_{\pm k} \hat{a}_{\pm k}) + 2 \frac{V_{xx}}{\hbar} B_{\pm k_p}^* B_{\pm k_p} \hat{B}_{\pm k} - 2 \frac{i}{\hbar} B_{\pm k_p}^*(t) \int_{-\infty}^{t} dt' F_{\pm\pm}(t - t') \hat{B}_{\pm k}(t') B_{\pm k_p}(t') \right). \tag{57}
\]

Equations (56) are the main result of this paper. They can be considered the starting point for the microscopic description of quantum optical effects in SMCs. These equations extend the usual semiclassical description of Coulomb interaction effects, in terms of a mean-field term plus a genuine non-instantaneous four-particle correlation, to quantum optical effects. Analogous equations can be obtained starting from an effective Hamiltonian describing excitons as interacting bosons \[10\]. The resulting equations (usually developed in a polariton basis) do not include correlation effects beyond Hartree-Fock. Moreover the interaction terms due to phase space filling differs from those obtaind within the present approach not based on an effective Hamiltonian.

Only the many-body electronic Hamiltonian, the intracavity-photon Hamiltonian and the Hamiltonian describing their mutual interaction have been taken into account. Losses through mirrors, decoherence and noise due to environment interactions as well as applications of this theoretical framework will be addressed in another paper of ours \[23\].

VI. CONCLUSION

In this paper we set a dynamics controlled truncation scheme approach to nonlinear optical processes in cavity embedded semiconductor QWs without any assumption on the quantum statistics of the excitons involved. This approach represents the starting point for the microscopic analysis to quantum optics experiments in the strong coupling regime.
We presented a systematic theory of Coulomb-induced correlation effects in the nonlinear optical processes in SMCs. We end up with dynamical equations for exciton and photon operators which extend the usual semiclassical description of Coulomb interaction effects, in terms of a mean-field term plus a genuine non-instantaneous four-particle correlation, to quantum optical effects. The proper inclusion of the detrimental environment interactions as well as applications of the present theoretical scheme will be presented in another paper of ours [23].


APPENDIX A: THE EQUATION OF MOTION AT ANY ORDER

The equation of motion for the operators in (10), under the Hamiltonian $\hat{H} = \hat{H}_e + \hat{H}_c + \hat{H}_f + \hat{H}_p$ reads:

$$\frac{d}{dt} \left( \hat{X}_{\alpha;\beta} \hat{Y}_{n;\mu} \right) = -i(\omega_M - \omega_N) + \sum_{i=1}^{n} \omega_{ki}^{\alpha} - \sum_{j=1}^{n} \omega_{kj}^{\alpha} \left( \hat{X}_{\alpha;\beta} \hat{Y}_{n;\mu} \right) +$$

$$+ \hat{X}_{\alpha;\beta} \left( \delta_{m,1} t_{e} \frac{E_{e}}{\hbar} \hat{Y}_{n;0} + \delta_{n,1} t_{c} \frac{E_{c}}{\hbar} \hat{Y}_{0;\mu} \right) - \hat{X}_{\alpha;\beta} \sum_{k} t_{e} \left( \delta_{m,0} \frac{E_{k}}{\hbar} \hat{Y}_{n;1 \kappa} + \delta_{n,0} \frac{E_{k}}{\hbar} \hat{Y}_{1 \kappa;\mu} \right) +$$

$$+ \sum_{k\nu} t_{e} \hat{X}_{\alpha;\beta} \left[ \Theta(n - 2) \langle (n - 1) \nu | \hat{\alpha}_{k} | (n - 1) \nu \rangle \frac{E_{k}}{\hbar} \hat{Y}_{n;\lambda;\nu} \right. -$$

$$\left. \Theta(n - 2) \langle (n - 1) \nu | \hat{\alpha}_{k} | (n + 1) \nu \rangle \frac{E_{k}}{\hbar} \hat{Y}_{n;\lambda;\nu} \right] +$$

$$+ i \frac{\delta_{M,0} \delta_{\beta,0} \delta_{m,1}}{\hbar} \sum_{n} V_{n\mu}^{*} \hat{X}_{\alpha;1 n \mu} \hat{Y}_{n;\lambda;0} - i \frac{\delta_{N,0} \delta_{\alpha,0} \delta_{n,1}}{\hbar} \sum_{n} V_{n\lambda} \hat{X}_{1 n \lambda;\beta} \hat{Y}_{0;\mu} -$$

$$- \frac{i}{\hbar} \delta_{N,1} \delta_{\alpha,0} \delta_{m,0} V_{\alpha}^{*} \hat{X}_{0;\beta} \hat{Y}_{1 \kappa;\mu} + \frac{i}{\hbar} \delta_{M,1} \delta_{m,0} \delta_{\mu,0} V_{\beta} \hat{X}_{\alpha;0} \hat{Y}_{n,1 \kappa} +$$

$$+ i \frac{\delta_{m,1} \Theta(n - 2)}{\hbar} \sum_{n \delta} V_{n\lambda} \langle (n + 1) \eta | \hat{\beta}_{n\delta} | (n + 1) \eta \rangle \hat{X}_{(n + 1) \eta;\beta} \hat{Y}_{0;\mu} -$$

$$- i \frac{\delta_{n,1} \Theta(n - 2)}{\hbar} \sum_{n \eta} V_{n\lambda} \langle (n + 1) \eta | \hat{\beta}_{n\delta} | (n + 1) \eta \rangle \hat{X}_{(n + 1) \eta;\beta} \hat{Y}_{0;\mu} +$$

$$+ i \frac{\delta_{m,0} \Theta(n - 2)}{\hbar} \sum_{n \delta} V_{n\lambda} \langle (n + 1) \eta | \hat{\beta}_{n\delta} | (n + 1) \eta \rangle \hat{X}_{(n + 1) \eta;\beta} \hat{Y}_{1 \kappa;\mu} +$$

$$+ i \frac{\delta_{m,0} \Theta(n - 2)}{\hbar} \sum_{n \kappa \nu} V_{n\lambda} \langle (m + 1) \nu | \hat{\alpha}_{n \kappa} | (m + 1) \nu \rangle \hat{X}_{\alpha;1 n \kappa} \hat{Y}_{n,\lambda;\nu} -$$

$$+ i \frac{\delta_{N,0} \Theta(n - 2)}{\hbar} \sum_{n \kappa \gamma} V_{n\lambda} \langle (n - 1) \gamma | \hat{\alpha}_{n \kappa} | (n - 1) \gamma \rangle \hat{X}_{1 n \kappa;\beta} \hat{Y}_{(n - 1) \gamma;\mu} -$$

$$- i \frac{\delta_{N,1} \Theta(n - 2)}{\hbar} \sum_{n \kappa \gamma} V_{n\lambda} \langle (n - 1) \gamma | \hat{\alpha}_{n \kappa} | (n - 1) \gamma \rangle \hat{X}_{1 n \kappa;\beta} \hat{Y}_{(n - 1) \gamma;\mu} +$$

$$+ i \frac{\delta_{M,1} \Theta(n - 2)}{\hbar} \sum_{n \nu} \langle (m + 1) \nu | \hat{\alpha}_{n \nu} | (m + 1) \nu \rangle \hat{X}_{\alpha;0 \nu} \hat{Y}_{n,\lambda;\nu} +$$

$$+ i \frac{\delta_{M,1} \Theta(n - 2)}{\hbar} \sum_{n \nu} \langle (m + 1) \nu | \hat{\alpha}_{n \nu} | (m + 1) \nu \rangle \hat{X}_{\alpha;0 \nu} \hat{Y}_{n,\lambda;\nu} +$$

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\[ + \frac{i}{\hbar} \sum_{\vec{n}_k} \sum_{\nu} \left[ V_{\vec{n}_k}^* \left( \Theta(M - 1) \Theta(m - 2) \langle M \beta | \hat{B}_{\vec{n}_k} | (M + 1) \delta \right) \right. \\
\left. \langle m \mu | \hat{a}_{\vec{k}}^\dagger | (m - 1) \nu \rangle \hat{X}_{N \alpha; (M + 1) \delta} \hat{Y}_{n \lambda; (m - 1) \nu} - \right. \\
\left. \Theta(N - 2) \Theta(n - 1) \langle (N - 1) \delta | \hat{B}_{\vec{n}_k} | N \alpha \rangle \right) \\
\left. \langle (n + 1) \nu | \hat{a}_{\vec{k}}^\dagger | n \lambda \rangle \hat{X}_{(N - 1) \delta; M \beta} \hat{Y}_{(n + 1) \nu; m \mu} + \right) \\
- \frac{i}{\hbar} V_{\vec{n}_k} \left( \Theta(N - 1) \Theta(n - 2) \langle (N + 1) \delta | \hat{B}_{\vec{n}_k}^\dagger | N \alpha \rangle \right) \\
\left. \langle (n - 1) \nu | \hat{a}_{\vec{k}} | n \lambda \rangle \hat{X}_{(N + 1) \delta; M \beta} \hat{Y}_{(n - 1) \nu; m \mu} - \right. \\
\left. \Theta(M - 2) \Theta(m - 1) \langle M \beta | \hat{B}_{\vec{n}_k}^\dagger | (M - 1) \delta \rangle \right. \\
\left. \langle m \mu | \hat{a}_{\vec{k}} | (m + 1) \nu \rangle \hat{X}_{N \alpha; (M - 1) \delta} \hat{Y}_{n \lambda; (m + 1) \nu} \right]. \tag{A1} \]

Here \( \Theta(x) \) is the Heaviside function equal to 1 for positive argument and zero otherwise.

**APPENDIX B: N \textit{E}H \textit{P}AIR \textit{A}GGREGATES**

We start from the usual model for the electronic Hamiltonian of a direct two-band semiconductor \[15, 24\]. It is obtained from the many-body Hamiltonian of the interacting electron system in a lattice, keeping explicitly only those terms in the Coulomb interaction preserving the number of electrons in a given band and can be expressed as

\[ \hat{H}_\text{e} = \hat{H}_0 + \hat{V}_\text{Coul}. \tag{B1} \]

It comprises the single-particle Hamiltonian terms for electrons in conduction band and holes in valence band (here \( k \equiv (k, \sigma) \) and \( \hat{c}_{\sigma,k} (\hat{d}_{\sigma,k}) \) annihilates an electron (a hole)):

\[ \hat{H}_0 = \sum_k E_{c,k} \hat{c}_{k}^\dagger \hat{c}_k + \sum_k E_{h,k} \hat{d}_{k}^\dagger \hat{d}_k, \tag{B2} \]

and the Coulomb interaction term of three contributions, the two repulsive electron-electron (e-e) and hole-hole (h-h) terms and the attractive (e-h) one:

\[ \hat{V}_\text{Coul} = - \frac{1}{2} \sum_{q \neq 0} \sum_{\sigma,k,\sigma',k'} \hat{c}_{\sigma,k+q}^\dagger \hat{c}_{\sigma',k-q}^\dagger \hat{c}_{\sigma,k} + \frac{1}{2} \sum_{q \neq 0} \sum_{\sigma,k,\sigma',k'} \hat{d}_{\sigma,k+q}^\dagger \hat{d}_{\sigma',k-q}^\dagger \hat{d}_{\sigma',k} \hat{d}_{\sigma,k} - \sum_{q \neq 0} \sum_{\sigma,k,\sigma',k'} \hat{c}_{\sigma,k+q}^\dagger \hat{d}_{\sigma',k-q}^\dagger \hat{d}_{\sigma',k} \hat{c}_{\sigma,k}. \tag{B3} \]

A many-body interacting state is usually very different from a product state, however a common way to express the former is by a superposition of uncorrelated product states.
The physical picture that arises out of it expresses the *dressing* the interaction performs over a set of noninteracting particles. The general many-body Schrödinger equation for this Coulomb-correlated system is

\[
\hat{H}_e \left| \Psi \right\rangle = (\hat{H}_0 + \hat{V}_{\text{Coul}}) \left| \Psi \right\rangle = E \left| \Psi \right\rangle ,
\]

(B4)

with \( \left| \Psi \right\rangle \) the global interacting many-body state of the whole Fock space and \( E \) its corresponding energy. The system Hamiltonian commutes with the total-number operators for electron and holes, i.e. \( \hat{N}_e = \sum_k \hat{c}_k^\dagger \hat{c}_k \) and \( \hat{N}_h = \sum_k \hat{d}_k^\dagger \hat{d}_k \). Therefore the state \( \left| \Psi \right\rangle \) may be build up corresponding on a given number of electrons and of holes. Moreover, because we shall consider the case of intrinsic semiconductors materials where \( N_e = N_h = N \), the good quantum number for the Schrödinger equation (B4) is the total number of electron-hole pairs \( N \), explicitly

\[
\hat{H}_e \left| N\alpha \right\rangle = E_{N\alpha} \left| N\alpha \right\rangle ,
\]

(B5)

where \( \alpha \) is the whole set of proper quantum numbers needed to specify univocally the many-body state.

For any given number \( N \) of electron-hole pairs, the product-state set, built up from the single-particle states \( \{ \left| Na \right\rangle \} \) eigenstates of the noninteracting carrier Hamiltonian \( \hat{H}_0 \), is a natural complete basis of the N-pair sector of the global Fock space:

\[
\hat{H}_0 \left| Na \right\rangle = \epsilon_{Na} \left| Na \right\rangle ,
\]

(B6)

where \( N \) identifies the N-pair subspace and \( a \) is a compact form for all the single particle indexes, i.e. \( a \equiv j_{e1}, j_{e2}, ..., j_{eN}; j_{h1}, j_{h2}, ..., j_{hN} \). Indeed

\[
\left| N, a \right\rangle = \bigotimes_{n=1}^{N} \hat{c}_{j_{en}}^\dagger \hat{d}_{j_{hn}}^\dagger \left| 0, 0 \right\rangle \quad \text{and} \quad \epsilon_{Na} = \sum_{n=1}^{N} (\epsilon_{j_{en}} + \epsilon_{j_{hn}}) .
\]

(B7)

Being a complete orthonormal basis for the N-pair subspace we may expand the many-body state \( \left| N, \alpha \right\rangle \) over it, it yields

\[
\left| N\alpha \right\rangle = \sum_a U_a^{N\alpha} \left| Na \right\rangle .
\]

(B8)

It is only a matter of calculation to show that \( U_a^{N\alpha} \) is nothing but the envelope function of the N-pair aggregate, solution of the corresponding secular equation. Indeed the eigenvalue problem (B5) is transformed into:

\[
\sum_{a'} \left( \langle Na | \hat{H}_e | Na' \rangle - E_{N\alpha} \delta_{a,a'} \right) U_{a'}^{N\alpha} = 0 .
\]

(B9)
Namely N=1 leads to the exciton secular equation, whereas N=2 represents the biexciton (two pairs) Coulomb problem.

In order to be clearer we shall propose in details the N=1 exciton calculation. We shall work in the direct lattice \( \mathbf{r} \leftrightarrow \mathbf{r}_i \) (the former is a continuous variable whereas the latter is a point in the 3D lattice). Using the general mapping \[ \sum \mathbf{r}_i \leftrightarrow (1/v_0) \int d^3 \mathbf{r}, \]
\( \delta(\mathbf{r} - \mathbf{r}') = (\delta_{\mathbf{r}_i, \mathbf{r}_j}/v_0), \)
here \( v_0 \) is the unit cell volume and for simplicity the spin selection rules for the optically active states has been already taken into account, \( (B9) \) reads

\[ \sum_{\mathbf{r}_e, \mathbf{r}_h} \left( \langle \mathbf{r}_e, \mathbf{r}_h \mid \hat{H}_e \mid \mathbf{r}'_e, \mathbf{r}'_h \rangle - E_{\sigma \mathbf{k}} \delta_{\mathbf{r}_e, \mathbf{r}_h, \mathbf{r}'_e, \mathbf{r}'_h} \right) U_{a}^{\sigma} (\mathbf{r}'_e, \mathbf{r}'_h) = 0, \] \( (B10) \)

with

\[ \langle \mathbf{r}_e, \mathbf{r}_h \mid \hat{H}_e \mid \mathbf{r}'_e, \mathbf{r}'_h \rangle = \left( -\frac{\hbar^2}{2m_e} \nabla^2_{\mathbf{r}_e} - \frac{\hbar^2}{2m_h} \nabla^2_{\mathbf{r}_h} - \frac{e^2}{\varepsilon_e |\mathbf{r}_e - \mathbf{r}_h|} + V(\mathbf{r}_e, \mathbf{r}_h) \right) \delta_{\mathbf{r}_e, \mathbf{r}_h, \mathbf{r}'_e, \mathbf{r}'_h}, \] \( (B11) \)

here \( V(\mathbf{r}_e, \mathbf{r}_h) \) represents all the additional potential, e.g. those of the heterostructures or those of disorder effects, \( V(\mathbf{r}_e, \mathbf{r}_h) = V^e(z_e) + V^h(z_h) \). Typically, the energy difference between the lowest QW subband level and the first excited one (at least a few meV) is much larger than the Coulomb interaction between particles (a few meV). As a consequence, at least at low temperatures, particles are confined at the lowest quantization level and the (possible) distorsion of the wave function due to the Coulomb-activated admixture of different subbands can be safely neglected. In some extent, then, the particle wave function dependence along the growth (say \( z \)) direction can be factorized out and the dynamics becomes essentially two-dimensional. However, a purely 2D approximation for excitons would miss important effects of the geometrical QWs parameters on the binding energy and would not be able to account for the interaction with a 3D continuum environment of surrounding modes (e.g. acoustic phonon modes in heterostructures with alloy lattice constant in close proximity \[ \text{[34]} \]). In addition in QWs, light and heavy holes in valence band are split off in energy. Assuming that this splitting is much larger than kinetic energies of all the involved particles and, as well, much larger than the interaction between them, we shall consider only heavy hole states as occupied.

In Eq. \( (B10) \) the 3D Coulomb interaction prevents form factorizing into (free) in-plane and confined directions. Nevertheless if we assume that the quantization energy along \( z \) is
much larger than the Coulomb energy, at leading order we can factorize out the $z$-dependence
\[
\left( -\frac{\hbar^2}{2m_e} \frac{d^2}{dz_e^2} + V_e(z_e) - \frac{\hbar^2}{2m_h} \frac{d^2}{dz_h^2} + V_h(z_h) \right) U^\alpha(r_e, r_h) = E^z U^\alpha(r_e, r_h).
\] (B12)

It means we are solving our secular equation with solutions built up as linear combination of $F^\alpha_{n_c,n_e,a}(r_\parallel, r_\perp)c_{n_c}(z_e)v_{n_e}(z_h)$, with $r = (r_\parallel, z)$. Equation (B12) expresses the lack of translational symmetry along the growth $z$ direction, thus single particle states experience confinement and two additional QW subband quantum numbers $n_v, n_c$ (for valence and conduction states respectively) appear. We still leave $a$ as a reminder that new possible indexes could still arise in due course.

Projecting Eq. (B10) on these confined states we end up with an effective Schrödinger equation in the plane
\[
\left( -\frac{\hbar^2}{2m_e} \nabla^2_{r_e} - \frac{\hbar^2}{2m_h} \nabla^2_{r_h} - U_{n_c,n_v;n_c',n_v'}(|r_\parallel - r_\parallel|) \right) F^\alpha_{n_c,n_v,a}(r_\parallel, r_\parallel) =
\]
\[
= (E_\alpha - E_{n_c}^z - E_{n_v}^z) F^\alpha_{n_c,n_v,a}(r_\parallel, r_\parallel),
\] (B13)

with
\[
U_{n_c,n_v;n_c',n_v'}(|r_\parallel - r_\parallel|) = \int dz_e \int dz_h \frac{e^2}{\varepsilon_r \sqrt{|r_\parallel - r_\parallel|^2 + (z_e - z_h)^2}} c_{n_c}(z_e)c_{n_c'}(z_e)v_{n_v}(z_h)v_{n_v'}(z_h).
\] (B14)

For what already stated, we shall consider only the lowest confined subband levels, then the resulting effective in-plane secular equation becomes
\[
\left( -\frac{\hbar^2}{2m_e} \nabla^2_{r_e} - \frac{\hbar^2}{2m_h} \nabla^2_{r_h} - \int dz_e \int dz_h \frac{e^2}{\varepsilon_r \sqrt{|r_\parallel - r_\parallel|^2 + (z_e - z_h)^2}} |c_{n_c}(z_e)|^2 |v_{n_v}(z_h)|^2 \right) F^\alpha_a(r_\parallel, r_\parallel) =
\]
\[
= (E_\alpha - E_{n_c}^z - E_{n_v}^z) F^\alpha_a(r_\parallel, r_\parallel),
\] (B15)

with the product exciton envelope function $U^\alpha(r_e, r_h) = F^\alpha_a(r_\parallel, r_\parallel)c(z_e)v(z_h)$. Equation (B15) is solvable by separation of variables once we employ a coordinate transformation into center of mass (CM) $\mathbf{R} = (m_e r_e + m_h r_h)/(m_e + m_h)$ and relative $\rho = (r_e - r_h)$ exciton coordinates. It reads
\[
\left( -\frac{\hbar^2}{2M} \nabla^2_{\mathbf{R}} - \frac{\hbar^2}{2\mu} \nabla^2_{\rho} - U(\rho) \right) F^\alpha_a(\mathbf{R}, \rho) = EF^\alpha_a(\mathbf{R}, \rho),
\] (B16)
with a solution we can arrange as $F^α_a(R, ρ) = \frac{e^{iKR}}{\sqrt{A}} W^α_a(ρ)$ the latter solution of the relative hydrogen-like 2D problem.

Eventually, in real-space representation, we have our exciton wave function with total in-plane CM wave vector $K$ ($A$ is the in-plane quantization surface in the free directions) which reads

$$| nσK⟩ = \frac{v_0}{\sqrt{A}} \sum_{r_{e,h}} e^{ikR} W_{nσ}(ρ)c(z_ρ) v(z_h) a^+_c r, r_h a^+_v | 0⟩,$$

(B17)

being $a^+_c, a^+_v$ creation (annihilation) operator of the conduction- or valence-band electron in the Wannier representation and $r_{e,h} = (r_h, z_h)$ are to be considered coordinates of the direct lattice, $| 0⟩$ is the crystal ground state.

When e.g. exploring the exciton-phonon interaction, it is useful to express exciton states in reciprocal space. With the usual transformation to Bloch representation, ($N = v_0 A L$ is the number of unit cells and $L$ is the quantization dimension along the confined direction, $ν = c, v$),

$$\hat{a}_ν(r, z) = \frac{1}{\sqrt{N}} \sum_{k, k_z} e^{ikR} a^+_ν(k, k_z),$$

(B18)

one obtains:

$$| nσK⟩ = \sum_{k, k_z} δ_{K-k'} \left( \frac{1}{\sqrt{A L}} \int dp \int dz_ρ \int dz_h W_{nσ}(ρ)c(z_ρ) v(z_h) e^{-iρ(η_h k + η_c k')} e^{-ik_z z_ρ} e^{-ik_z z_h} \right)$$

$$a^+_c(k, k_z) a^+_v(k', k_z') | 0⟩.$$

(B19)

In order to end up with a form as much as possible in analogy with its bulk counterpart we shall define CM and relative coordinates even in the reciprocal lattice:

$$\begin{align*}
K &= k - k' \\
k_r &= η_h k + η_c k' \\
k' &= k_r - η_h K
\end{align*}$$

(B20)

It becomes

$$| nσK⟩ = \sum_{K, k_r} δ_{K-k'} \sum_{k_z, k_z'} \left( \frac{1}{\sqrt{A}} \int dp W_{nσ}(ρ) e^{-iρkr} \right) \left( \frac{1}{\sqrt{L}} \int dz_ρ c(z_ρ) e^{-ik_z z_ρ} \right)$$

$$\left( \frac{1}{\sqrt{L}} \int dz_h v(z_h) e^{-ik_z' z_h} \right) a^+_c(k_r + η_c K, k_z) a^+_v(k_r - η_h K, k_z') | 0⟩.$$

(B21)

Thus

$$| nσK⟩ = \sum_{k_r} \sum_{k_z, k_z'} φ^K_{nσ}, k_r, k_z, k_z' a^+_c(k_r + η_c K, k_z) a^+_v(k_r - η_h K, k_z') | 0⟩,$$

(B22)
or in the electron-hole picture ($\hat{a}_{v,k} = \hat{d}^{\dagger}_{-k}$ and $-k|_{el} = k|_{hole}$)

$$| n\sigma K \rangle = \sum_{k_r} \sum_{k_z,k_z'} \Phi_{n\sigma,k_r,k_z}^{K} u_{k_z'}^{h} u_{k_z}^{e} \epsilon_{(k_r+\eta_{k},k_z')} \hat{d}^{\dagger}_{(-k_r+\eta_{k},-k_z')} |0\rangle,$$

(B23)

with the relations in Eq. (B20) changed accordingly.

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[28] Inserting two photon identities at both sides \( \hat{a}_k = \sum_{n,\gamma} Y_{n,\gamma;n,\gamma} \hat{a}_k \sum_{m,\lambda} Y_{m,\lambda;m,\lambda} \), where \( n = \sum_{i=modes} n_i \) and \( m = \sum_{i=modes} m_i \), we have non-zero matrix elements only if \( n_i = m_i \forall i \neq k \) and \( n_k + 1 = m_k \). Thus it becomes \( \hat{a}_k = \sum_{(n_k,n'),\gamma}(\sqrt{n_k+1}Y_{n_k;k;(n_k+1)k}) \otimes Y_{n';\gamma;n'\gamma} \), where \( n' \) stands for the string \( (n_1,n_2,...) \) without the \( k \)-th entry. When we make the one photon addendum explicit, we end up with \( Y_{0;1k} \).

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