Dynamics-controlled truncation scheme for quantum optics and nonlinear dynamics in semiconductor microcavities

Dynamics-controlled truncation scheme for quantum optics and nonlinear dynamics in semiconductor microcavities

S. Portolan,[1,3,*] O. Di Stefano,[2] S. Savasta,[2] F. Rossi,[3] and R. Girlanda[2]
[1]*Institute of Theoretical Physics, Ecole Polytechnique Fédérale de Lausanne EPFL, CH-1015 Lausanne, Switzerland*
[2]*Dipartimento di Fisica della Materia e Tecnologie Fisiche Avanzate, Università di Messina Salita Sperone 31, I-98166 Messina, Italy*
[3]*Dipartimento di Fisica, Politecnico di Torino, Corso Duca degli Abruzzi 24, I-10129 Torino, Italy*
(Received 16 October 2007; revised manuscript received 13 February 2008; published 5 May 2008)

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 microscopic treatment of nonlinear parametric processes in semiconductor microcavities 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, which was used in the semiclassical context, which, once applied to four-wave mixing in quantum wells, allowed us to understand a wide range of observed phenomena. 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 noninstantaneous four-particle correlation to quantum optical effects.

DOI: 10.1103/PhysRevB.77.195305 PACS number(s): 42.50.−p, 42.65.−k, 71.35.−y, 71.36.+c

I. INTRODUCTION

Since the early 1970s[1] researchers have been exploring the possible realization of semiconductor-based heterostructures, which were devised according to the principles of quantum mechanics. The development of sophisticated growth techniques started a revolution in semiconductor physics, which was 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[2] 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 and compact sources of nonclassical photon states, which enables efficient coupling into optical fibers 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.[3] Demonstrations of parametric amplification and parametric emission in semiconductor microcavities (SMCs) with embedded QWs,[4–6] 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.[7–10] In 2004, squeezed light generation in SMCs in the strong-coupling regime was demonstrated.[11] In 2005, an experiment probing quantum correlations of (parametrically emitted) cavity polaritons by exploiting quantum complementarity was proposed and realized.[8] 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 experiments as well as coherent control, manipulation, creation, and measurement of nonclassical states.[8,12–14] 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 properly include the effects of noise and dephasing induced by the electron-phonon interaction and the other environment channels.[15] 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 for in the latter, the optical response is well described by independent transitions between atomic levels and the nonlinear dynamics is governed only by saturation effects, which are mainly due to the balance of populations between different levels.

The dynamics controlled truncation scheme[16] (DCTS) provides a (widely adopted) starting point for the microscopic theory of the light-matter interaction effects beyond mean-field,[12] supplying a consistent and precise way to stop the infinite hierarchy of higher-order correlations which always appear 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 exploting 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 entirely contained in a retarded memory function.[14] In 1996, the DCTS was extended in order to include in the description the quantization of the electromagnetic field and polariton effects.[17] This extension has been applied to the study of quantum optical phenomena in semiconductors and it was exploited to predict polariton entanglement.[10] 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 explicitly included biexciton states. The structure of those equations did not 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[14] which, once applied to four-wave mixing in QWs, allowed us to understand a wide range of observed phenomena. We shall provide an effective description of the nonlinear parametric contributions beyond mean field in an exciton-exciton correlation fashion. In particular, we shall derive the coupled equations of motion for the excitonic polarization and the intracavity field. It shows a close analogy to the corresponding equation, which describes the semiclassical (quantized electron system and classical light field) coherent $\chi^{(3)}$ response in a QW;[14] the main difference is that the (intracavity) light field here is regarded not as a driving external source but as a dynamical field.[18] This correspondence is a consequence of the linearization of quantum fluctuations in the nonlinear source term adopted here, namely, the standard linearization procedure of quantum correlations adopted for large systems.[19] 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 the expectation values of exciton and photon numbers or even higher-order correlators, which are key quantities for the investigation of coherence properties of quantum light.[8] This is the main motivation for the derivation of fully operatorial dynamical equations within such lowest-order nonlinear coherent response, which we shall address in this paper. The results presented here 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., Refs. 12, 14, 18, 20, and 21 and references therein. The proper inclusion of the detrimental environmental interaction, which is an important and compelling issue, is left for a detailed analysis in another paper of ours.[22]

In Sec. II, the generalities of the coupled system taken into account are exposed; here, all the ingredients, which contribute to the dynamics, are introduced and commented. The linear and the lowest nonlinear dynamics is the subject of Sec. III, whereas in Sec. IV, the operatorial equations of motion for exciton and intracavity photon operators are derived. We shall give the needed manipulations required in order to provide an effective description of the nonlinear parametric contributions beyond mean field in an exciton-exciton correlation fashion. Section V will be devoted to the strong coupling regime and the transformation of the optical response description in terms of interacting polaritons; we will consider its weaknesses and strength even in comparison with the existing literature. Finally, in Sec. VI, we shall summarize and draw some conclusions.

II. COUPLED SYSTEM

The system we have in mind is a semiconductor QW grown inside a semiconductor planar Fabry–Pérot 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 such as 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 twofold degenerate heavy valence subbands with $j_z=\pm 3/2$ and light lower energy subbands with $j_z=\pm 1/2$. The conduction band, which arises 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 spinlike 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 $\sigma=-(+)$ excite electrons with $j_z^e=+1/2$ ($j_z^e=-1/2$) and holes with $j_z^h=-3/2$ ($j_z^h=3/2$). We label the optically active eh pairs with the same polarization label of light generating them; e.g., $\sigma=+1$ indicates an eh pair with $j_z^e=-1/2$ and $j_z^h=3/2$.

We start from the usual model for the electronic Hamiltonian of semiconductors.[12,23] It is obtained from the many-body Hamiltonian of the interacting electron system in a lattice, which explicitly keeps 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

$$\hat{H}_e = \hat{H}_0 + \hat{V}_{\mathrm{Coul}} = \sum_{N\alpha} E_{N\alpha} |N\alpha\rangle\langle N\alpha|, \tag{1}$$

where the eigenstates of $\hat{H}_e$, with energies $E_{N\alpha}=\hbar\omega_{N\alpha}$, have been labeled 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 α which denotes the exciton energy level n, the in-

plane wave vector $\mathbf{k}$, and the spin index σ. 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. By 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,

$$|1n\sigma\mathbf{k}\rangle = \hat{B}^{\dagger}_{n\sigma\mathbf{k}}|N=0\rangle, \tag{2}$$

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

$$\hat{B}^{\dagger}_{n\sigma\mathbf{k}} = \sum_{\mathbf{k}'} \Phi^{\mathbf{k}}_{n\sigma\mathbf{k}'} \hat{c}^{\dagger}_{\sigma,\mathbf{k}'+\eta_e\mathbf{k}} \hat{d}^{\dagger}_{\sigma,-\mathbf{k}'+\eta_h\mathbf{k}}. \tag{3}$$

Here, $\Phi^{\mathbf{k}}_{n\sigma\mathbf{k}'}$ is the exciton wave function, with $\mathbf{k}$ as the total wave vector $\mathbf{k}=\mathbf{k}_e+\mathbf{k}_h$ and $\mathbf{k}'=\eta_h\mathbf{k}_e-\eta_e\mathbf{k}_h$, 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 $\hat{H}_e$,

$$\hat{H}_e|1n\sigma\mathbf{k}\rangle = \hbar\omega_{1n\sigma\mathbf{k}}|1n\sigma\mathbf{k}\rangle, \tag{4}$$

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

$$\sum_{k_e,k_h} (\langle k'_e,k'_h|\hat{H}_e|k_e,k_h\rangle - \hbar\omega_{1n\sigma\mathbf{k}}\delta_{k'_e k'_h,k_e k_h})\langle k_e,k_h|1n\sigma\mathbf{k}\rangle = 0 \tag{5}$$

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

$$|1n\sigma\mathbf{k}\rangle = \sum_{k_e,k_h} (\langle k_e,k_h|1n\sigma\mathbf{k}\rangle)|k_e,k_h\rangle, \tag{6}$$

the scalar products, which are the coefficients of this expansion, represent nothing, but the envelope function $\Phi^{\mathbf{k}}_{n,\sigma,\mathbf{k}'}$ of the excitonic aggregate, solution of the corresponding Schrödinger equation [Eq. (5)]. It describes the correlated *eh* relative motion in the k-space. In order to simplify the notation a bit, 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, which spans all the states with two *eh* pairs. It seems worth noting that the above description of *eh* complexes arises from the properties of quantum states, and once the system Hamiltonian is fixed, 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 $\lambda=(\mathbf{k}_1,\sigma_1;\ldots;\mathbf{k}_n,\sigma_n)$ specifies the wave vector and polarization σ of each photon. Here, we shall neglect the longitudinal-transverse splitting of polaritons,[24] which mainly originate 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 quasimode 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

$$\hat{H}_c = \sum_k \hbar\omega^c_k \hat{a}^{\dagger}_k \hat{a}_k, \tag{7}$$

with the operator $\hat{a}^{\dagger}_k$ which creates a photon state with energy $\hbar\omega^c_k = \hbar(\omega^2_{\mathrm{exc}} + v^2|\mathbf{k}|^2)^{1/2}$, where v is the velocity of light inside the cavity and $k=(\sigma,\mathbf{k})$. The coupling between the electron system and the cavity modes is given in the usual rotating wave approximation,[17,25]

$$\hat{H}_I = -\sum_{nk} V^*_{nk} \hat{a}^{\dagger}_k \hat{B}_{nk} + \mathrm{H.c.}, \tag{8}$$

where V_{nk} is the photon-exciton coupling coefficient enhanced by the presence of the cavity[26] set as $V_{nk} = \tilde{V}_\sigma \sqrt{A}\,\phi^*_{n,\sigma}(\mathbf{x}=0)$; the latter is the real-space exciton envelope function calculated in the origin, whereas A is the in-plane quantization surface, and $\tilde{V}_\sigma$ is proportional to the interband dipole matrix element. Modeling the loss through the cavity mirrors within the quasimode 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 nonzero expectation value of the ensemble. It can be shown[17,22] that for a coherent input beam, the driving of the cavity modes may be described by the model Hamiltonian,[17,22]

$$\hat{H}_p = it_c \sum_{\mathbf{k}} (E_{\mathbf{k}}\hat{a}^{\dagger}_{\mathbf{k}} - E^*_{\mathbf{k}}\hat{a}_{\mathbf{k}}), \tag{9}$$

where t_c determines the fraction of the field amplitude passing the cavity mirror, $E_{\mathbf{k}}$ ($E^*_{\mathbf{k}}$) is a $\mathbb{C}$–number, which describes 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 directly derive the expectation values of all the quantities at play. The dynamics is described by "transition" operators (known as generalized Hubbard operators),

$$\hat{X}_{N,\alpha;M,\beta} = |N,\alpha\rangle\langle M,\beta|,$$

$$\hat{Y}_{n,\lambda;m,\mu} = |n,\lambda\rangle\langle m,\mu|. \tag{10}$$

The fundamental point in the whole analysis is that, thanks to the form of the interaction Hamiltonian $\hat{H}_I$ and to the quasiparticle conservation the free Hamiltonians possess, we can use the so-called dynamics controlled truncation scheme, which states that we are facing a rather special model where the correlations have their origin only in the action of the electromagnetic field and thus the general theorem due to Axt and co-workers[16] holds. For our purpose, we will need its generalization in order to include the quantization of the electromagnetic field,[17] which reads

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

i.e., the expectation value of a zero to N-pair transition, even in the absence of cavity photons, 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

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

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

where in writing the photon expansion, we omitted all the states not belonging to the kth mode, which add up giving the identity in every Fock sector.[27]

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 by considering the equation of motion for the photon and exciton operators; once the expectation values are taken, 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 $\langle \hat{a}_k\rangle^{(1)} = \langle \hat{Y}_{0;1nk}\rangle^{(1)}$ and $\langle \hat{B}_{nk}\rangle^{(1)} = \langle \hat{X}_{0;1nk}\rangle^{(1)}$ reads

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

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

In these equations, $\bar{\omega}_k^c = \omega_k^c - i\gamma_k$, where γ_k is the cavity damping; analogously, we set $\bar{\omega}_{1nk} = \omega_{1nk} - i\Gamma_x$ for excitons (1 *eh* pairs) and $\bar{\omega}_{2\beta} = \omega_{2\beta} - i\Gamma_{xx}$ for biexcitons (two *eh* pairs). The dynamics up to the third order is a little bit more complex. We shall extensively use Eq. (A1) (in the following the superscript $+(n)$ stands for "up to" nth 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

$$\frac{d}{dt}\langle \hat{a}_k\rangle^{+(3)} = -i\bar{\omega}_k^c\langle \hat{a}_k\rangle^{+(3)} + i\sum_n \frac{V_{nk}^*}{\hbar}\langle \hat{B}_{nk}\rangle^{+(3)} + t_c\frac{E_k}{\hbar}, \tag{17}$$

$$\frac{d}{dt}\langle \hat{B}_{nk}\rangle^{+(3)} = -i\bar{\omega}_{1nk}\langle \hat{B}_{nk}\rangle^{+(3)} + i\frac{V_{nk}}{\hbar}\langle \hat{a}_k\rangle^{+(3)} - \frac{i}{\hbar}\sum_{\tilde{n}\tilde{k}}\left[\sum_{n'k',n''k''} C_{\tilde{n}\tilde{k},nk}^{n'k',n''k''}\langle \hat{X}_{1\tilde{n}\tilde{k};1n''k''}\hat{Y}_{0;1k'}\rangle^{(3)} + \sum_\beta c_{nk;\tilde{n}\tilde{k};\beta}\langle \hat{X}_{1\tilde{n}\tilde{k};2\beta}\rangle^{(3)}\right], \tag{18}$$

with

$$C_{\tilde{n}\tilde{k},nk}^{n'k',n''k''} = V_{n'k'}\langle 1\tilde{n}\tilde{k}|\delta_{(n'k');(nk)} - [\hat{B}_{nk},\hat{B}_{n'k'}^\dagger]|1n''k''\rangle, \tag{19}$$

$$c_{nk;\tilde{n}\tilde{k};\beta} = \hbar(\omega_{2\beta} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk})\langle 1\tilde{n}\tilde{k}|\hat{B}_{nk}|2\beta\rangle, \tag{20}$$

in analogy with the equations in Ref. 17 (see also Ref. 14). The resulting equation of motion for the lowest-order biexciton amplitude is

$$\frac{d}{dt}\langle \hat{X}_{0;2\beta}\rangle^{(2)} = -i\bar{\omega}_{2\beta}\langle \hat{X}_{0;2\beta}\rangle^{(2)} + \frac{i}{\hbar}\sum_{n'k';n''k''} V_{n'k'}\langle 2\beta|\hat{B}_{n'k'}^\dagger|1n''k''\rangle \times\langle \hat{X}_{0,1n''k''}\hat{Y}_{0,1k'}\rangle^{(2)}. \tag{21}$$

IV. PARAMETRIC PHOTOLUMINESCENCE: TOWARD SEMICONDUCTOR QUANTUM OPTICS

Entanglement is one of the key features of quantum information and communication technology.[2] 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. Two-photon parametric processes in semiconductor or in SMCs (known also as parametric emission) is a $\chi^{(3)}$ process analogous to $\chi^{(2)}$ parametric down-conversion. In the latter, a pair of photons is down-converted into a pair of lower-energy photons with $\omega_1+\omega_2=\omega_p$, whereas in $\chi^{(3)}$ parametric emission, a pair of pump polaritons scatters into a different pair as $2\omega_p=\omega_1+\omega_2$. 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 was proposed and realized.[8] Within the DCTS, we shall employ Eqs. (17), (18), and (21) in operatorial form, provided that all equations become fully significant as soon as the expectation value quantities that we shall work out would lie within the consistent perturbative DCTS order that we set from the beginning.[25] They read

$$\frac{d}{dt}\hat{a}_k = -i\omega_k^c\hat{a}_k + i\sum_n \frac{V_{nk}^*}{\hbar}\hat{B}_{nk} + t_c\frac{E_k}{\hbar}, \tag{22}$$

$$\frac{d}{dt}\hat{B}_{nk} = -i\omega_{1nk}\hat{B}_{nk} + i\frac{V_{nk}}{\hbar}\hat{a}_k - \frac{i}{\hbar}\sum_{\tilde{n}\tilde{k}}\left[\sum_{n'k',n''k''} C_{\tilde{n}\tilde{k},nk}^{n'k',n''k''}\hat{X}_{1\tilde{n}\tilde{k},0}\hat{X}_{0,1n''k''}\hat{Y}_{0,1k'} + \sum_\beta c_{nk;\tilde{n}\tilde{k};\beta}\hat{X}_{1\tilde{n}\tilde{k},0}\hat{X}_{0,2\beta}\right]. \tag{23}$$

A. Phase-space filling term

The first nonlinear term directly depends on the exciton wave function and represents a phase-space filling (PSF) contribution due to the Pauli blocking of electrons. Its analysis allows us to obtain a clear link with existing literature and to introduce experimental relevant parameters, such as vacuum Rabi splitting and saturation density. It can be developed as follows:

$$\begin{aligned} C_{\tilde{n}\tilde{k},nk}^{n'k',n''k''} &= V_{n'k'}\langle 1\tilde{n}\tilde{\sigma}\tilde{\mathbf{k}}|\delta_{(n'k');(nk)} - [\hat{B}_{n\sigma\mathbf{k}},\hat{B}^\dagger_{n'\sigma'\mathbf{k}'}]|1n''\sigma''\mathbf{k}''\rangle \\ &= V_{n'k'}\delta_{\sigma,\sigma'}\left\{\sum_{\mathbf{q}}\Phi^{\mathbf{k}*}_{n\sigma\mathbf{q}}\Phi^{\mathbf{k}'}_{n'\sigma'[\mathbf{q}+\eta_h(\mathbf{k}'-\mathbf{k})]}\langle 1\tilde{n}\tilde{\sigma}\tilde{\mathbf{k}}|\hat{c}^\dagger_{\sigma',\mathbf{q}+\eta_h(\mathbf{k}'-\mathbf{k})+\eta_e\mathbf{k}'}c_{\sigma,\mathbf{q}+\eta_e\mathbf{k}}|1n''\sigma''\mathbf{k}''\rangle\right. \\ &\quad \left. + \sum_{\mathbf{q}}\Phi^{\mathbf{k}*}_{n\sigma\mathbf{q}}\Phi^{\mathbf{k}'}_{n'\sigma'[\mathbf{q}-\eta_e(\mathbf{k}'-\mathbf{k})]}\langle 1\tilde{n}\tilde{\sigma}\tilde{\mathbf{k}}|\hat{d}^\dagger_{\sigma',-\mathbf{q}+\eta_e(\mathbf{k}'-\mathbf{k})+\eta_h\mathbf{k}'}d_{\sigma,-\mathbf{q}+\eta_h\mathbf{k}}|1n''\sigma''\mathbf{k}''\rangle\right\} \\ &= V_{n'k'}\delta_{\sigma,\sigma'}\delta_{\mathbf{k}+\tilde{\mathbf{k}};\mathbf{k}'+\mathbf{k}''}\left\{\sum_{\mathbf{q}}\Phi^{\mathbf{k}*}_{n\sigma\mathbf{q}}\Phi^{\mathbf{k}'}_{n'\sigma'\mathbf{q}_0}\Phi^{\tilde{\mathbf{k}}*}_{\tilde{n}\tilde{\sigma}\mathbf{q}_1}\Phi^{\mathbf{k}''}_{n''\sigma''\mathbf{q}_2} + \sum_{\mathbf{q}}\Phi^{\mathbf{k}*}_{n\sigma\mathbf{q}}\Phi^{\mathbf{k}'}_{n'\sigma'\mathbf{q}_3}\Phi^{\tilde{\mathbf{k}}*}_{\tilde{n}\tilde{\sigma}\mathbf{q}_4}\Phi^{\mathbf{k}''}_{n''\sigma''\mathbf{q}_5}\right\}. \end{aligned} \tag{24}$$

The explicit expressions of $\mathbf{q}$'s are such that

$$\mathbf{q}_0 = \mathbf{q} + \eta_h(\mathbf{k}'-\mathbf{k}), \quad \mathbf{q}_3 = \mathbf{q} - \eta_e(\mathbf{k}'-\mathbf{k}),$$

$$\mathbf{q}_1 = \mathbf{q} - \mathbf{k}'' + \eta_h\tilde{\mathbf{k}} + \eta_e\mathbf{k}, \quad \mathbf{q}_4 = \mathbf{q} + \mathbf{k}'' - \eta_e\tilde{\mathbf{k}} - \eta_h\mathbf{k},$$

$$\mathbf{q}_2 = \mathbf{q} - \mathbf{k}'' + \eta_e\mathbf{k} + \eta_h\mathbf{k}'', \quad \mathbf{q}_5 = \mathbf{q} + \mathbf{k}'' - \eta_h\mathbf{k} - \eta_e\mathbf{k}''.$$

Thus, the nonlinear dynamics of Eq. (23) driven by $\hat{H}_I$ can be written as

$$\frac{d}{dt}\hat{B}_{n\sigma\mathbf{k}}|_{\hat{H}_I} = +i\frac{V_{n\sigma\mathbf{k}}}{\hbar}\hat{a}_{\sigma\mathbf{k}} - \frac{i}{\hbar}\sum_{\substack{n'n''\tilde{n}\\ \mathbf{k}'\mathbf{k}''\tilde{\mathbf{k}}}}\delta_{\mathbf{k}+\tilde{\mathbf{k}};\mathbf{k}'+\mathbf{k}''}\hat{X}_{1\tilde{n}\tilde{\sigma}\tilde{\mathbf{k}},0}\hat{X}_{0,1n''\sigma\mathbf{k}''}\hat{Y}_{0,1\sigma\mathbf{k}'}\tilde{V}^*_\sigma\left[\sum_{\mathbf{q}}\Phi^{\mathbf{k}*}_{n\sigma\mathbf{q}}\Phi^{\tilde{\mathbf{k}}*}_{\tilde{n}\sigma\mathbf{q}_1}\Phi^{\mathbf{k}''}_{n''\sigma\mathbf{q}_2} + \sum_{\mathbf{q}}\Phi^{\mathbf{k}*}_{n\sigma\mathbf{q}}\Phi^{\tilde{\mathbf{k}}*}_{\tilde{n}\sigma\mathbf{q}_4}\Phi^{\mathbf{k}''}_{n''\sigma\mathbf{q}_5}\right]. \tag{25}$$

We are interested in studying polaritonic effects in SMCs where the optical response mainly involves excitons, which belongs to the $1S$ band with wave vectors close to normal incidence, i.e., $|\mathbf{k}| \ll \frac{\pi}{a_x}$ (much lower than the inverse of the exciton Bohr radius). In this case, exciton relative wave functions are independent on spins as well as on the center of mass wave vector. They can be expressed as $\Phi_{\mathbf{q}} = \frac{1}{\sqrt{A}}\frac{\sqrt{2\pi}2a_x}{[1+(a_x|\mathbf{q}|)^2]^{3/2}}$, where a_x is the exciton Bohr radius (notice that $\Sigma_{\mathbf{q}=-\infty}^{\infty}|\Phi_{\mathbf{q}}|^2=1$). As a consequence, in the $\mathbf{k}$ range of

interest, the matrix elements entering the nonlinear terms are largely independent on the wave vectors and we could calculate them for normal incidence: $\tilde{\mathbf{k}} \simeq \mathbf{k}' \simeq \mathbf{k}'' \simeq \mathbf{k} \simeq 0$. From now on, whenever no excitonic level is specified, the $1S$ label is understood. It yields

$$\frac{d}{dt}\hat{B}_{\sigma\mathbf{k}}|_{\hat{H}_I} = +i\frac{V_{\sigma\mathbf{k}}}{\hbar}\hat{a}_{\sigma\mathbf{k}} - \frac{i}{\hbar}\frac{V}{n_{\mathrm{sat}}}\sum_{\mathbf{k}'\mathbf{k}''\tilde{\mathbf{k}}} \delta_{\mathbf{k}+\tilde{\mathbf{k}};\mathbf{k}'+\mathbf{k}''}\hat{X}_{1\sigma\tilde{\mathbf{k}},0}\hat{X}_{0,1\sigma\mathbf{k}''}\hat{Y}_{0,1\sigma\mathbf{k}'}, \tag{26}$$

The last manipulation follows from

$$O^{\mathrm{PSF}} = \sum_{\mathbf{q}} \Phi_{\mathbf{q}}^{*}\Phi_{\mathbf{q}}^{*}\Phi_{\mathbf{q}},$$

$$\tilde{V}_{\sigma}^{*}O^{\mathrm{PSF}} = \frac{V}{\sqrt{A}\phi^{*}(0)}O^{\mathrm{PSF}} = \frac{8}{7}\frac{\pi a_x^2}{A}V = \frac{1}{2}\frac{V}{n_{\mathrm{sat}}}, \tag{27}$$

where $n_{\mathrm{sat}} \doteq (7/16)(A/\pi a_x^2)$ is called saturation density and we have defined $V \doteq V_{\sigma} = \tilde{V}_{\sigma}\sqrt{A}\phi^{*}(0)$ because, for the range of $\mathbf{k}$ space of interest, the so-called vacuum Rabi splitting $V_{n\sigma\mathbf{k}}$ (Ref. 4) of the $1S$ excitonic resonance is essentially constant, as can be experimentally checked.

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_{sat} is extensively used.[6,18,20,28] The other most interesting reason is that we can directly compute this quantity. Indeed, the equation of motion for the exciton operator reads

$$\frac{d}{dt}\hat{B}_{\pm\mathbf{k}} = -i\omega_{\mathbf{k}}\hat{B}_{\pm\mathbf{k}} + i\frac{V}{\hbar}\hat{a}_{\pm\mathbf{k}} - \frac{i}{\hbar}\sum_{\tilde{\sigma}=\pm\tilde{\mathbf{k}},\beta} c_{\pm\mathbf{k};\tilde{\sigma}\tilde{\mathbf{k}};\beta}\hat{X}_{1\tilde{\sigma}\tilde{\mathbf{k}},0}\hat{X}_{0,2\beta} - \frac{i}{\hbar}2\frac{V}{\sqrt{A}\phi^{*}(0)}O^{\mathrm{PSF}}\sum_{\mathbf{k}'\mathbf{k}''\tilde{\mathbf{k}}} \delta_{\mathbf{k}+\tilde{\mathbf{k}};\mathbf{k}'+\mathbf{k}''}\hat{X}_{1\pm\tilde{\mathbf{k}},0}\hat{X}_{0,1\pm\mathbf{k}''}\hat{Y}_{0,1\pm\mathbf{k}'}.$$

By leaving apart the discrepancy between the order in the DCTS, we can compute the so-called *oscillator strength* (OS), which is defined as what multiplies the photon at $\mathbf{k}=0$,[29]

$$\mathrm{OS} = i\frac{V}{\hbar}\left(1 - \frac{2}{\sqrt{A}\phi^{*}(0)}O^{\mathrm{PSF}}[\hat{X}_{1\pm(\tilde{\mathbf{k}}=0),0}\hat{X}_{0,1\pm(\mathbf{k}''=0)}]\right). \tag{28}$$

The saturation density may be defined as the exciton density that makes the oscillator strength to be zero. We obtain

$$n_{\mathrm{sat}} = \left(\frac{2}{\sqrt{A}\phi^{*}(0)}O^{\mathrm{PSF}}\right)^{-1} = \frac{A}{\pi a_x^2}\frac{7}{16}. \tag{29}$$

B. Coulomb-induced terms

Since we want to treat coherent optical processes, it is possible to manipulate further the parametric contributions. Since we are addressing a coherent optical response, 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 by considering the nonlinear term as mainly originating from coherent contributions. In the following, we will replace quantum operators at $\mathbf{k}=\mathbf{k}_p$ with classical amplitudes ($\mathbb{C}$–numbers). From the point of view of quantum effects, this approximation implies that nonclassical correlations are taken into account up to the lowest order, namely, the standard linearization procedure of quantum correlations adopted for large systems.[19]

The most common setup for parametric emission is the one where a single coherent pump feed resonantly excites the structure at a given energy and wave vector $\mathbf{k}_p$. The generalization to multipump setup is straightforward. In order to be more *specific*, we shall explicitly derive the case of input light beams activating only the $1S$ exciton sector with all the same circularly (e.g., σ^{+}) polarization, thus excluding the coherent excitation of bound two-pair coherences (biexciton) which are mainly responsible for polarization mixing.[14] The latter situation can be realized, as instance, as soon as the biexciton resonance has been carefully tailored off-resonance with respect to the characteristic energies of the states involved in the parametric scattering.[30] We shall show that under this approximation, we end up with a set of coupled equations analogous to those obtained in the semiclassical framework of coherent $\chi^{(3)}$ response (quantized electron system and classical light field) in a QW[14]; the main difference is that the (intracavity) light field here is regarded not as a driving external source but as a dynamical field.[18] Nevertheless, completely different results can be obtained for exciton or photon number expectation values or for higher-order correlation functions.[10,25] 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. In addition, having a precise setup 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. Starting from the Heisenberg equations for the exciton and photon operators with terms providing only lowest-order nonlinear response (in the input light field) [Eqs. (22) and (23)], we shall retain only the dominant contributions, namely, those containing the semiclassical pump amplitude at k_p twice, thus focusing on the "direct" pump-induced nonlinear parametric scattering processes. It reads

$$\frac{d}{dt}\hat{B}_{\pm\mathbf{k}} = -i\omega_{\mathbf{k}}\hat{B}_{\pm\mathbf{k}} + i\frac{V}{\hbar}\hat{a}_{\pm\mathbf{k}} - \frac{i}{\hbar}\frac{V}{n_{\mathrm{sat}}}\sum_{\tilde{\mathbf{k}},\mathbf{k}',\mathbf{k}''} \delta_{\mathbf{k}+\tilde{\mathbf{k}},\mathbf{k}'+\mathbf{k}''}\hat{X}_{1\pm\tilde{\mathbf{k}},0}\hat{X}_{0,1\pm\mathbf{k}''}\hat{Y}_{0;1\pm\mathbf{k}'}(\delta_{\mathbf{k}'',\mathbf{k}_p}\delta_{\mathbf{k}',\mathbf{k}_p} + \delta_{\tilde{\mathbf{k}},\mathbf{k}_p}\delta_{\mathbf{k}'',\mathbf{k}_p} + \delta_{\tilde{\mathbf{k}},\mathbf{k}_p}\delta_{\mathbf{k}',\mathbf{k}_p})$$

$$-\frac{i}{\hbar}\sum_{\tilde{\sigma}\tilde{\mathbf{k}},\sigma_\beta\mathbf{k}_\beta}\hbar(\omega_{2\mathbf{k}_\beta}-\omega_{1\tilde{\mathbf{k}}}-\omega_{1\mathbf{k}})\langle 1\tilde{\sigma}\tilde{\mathbf{k}}|\hat{B}_{\pm\mathbf{k}}|2\sigma_\beta\mathbf{k}_\beta\rangle\hat{X}_{1\tilde{\sigma}\tilde{\mathbf{k}},0}\hat{X}_{0,2\sigma_\beta\mathbf{k}_\beta}(\delta_{k_\beta,2k_p}+\delta_{\tilde{k},k_p}\delta_{k_\beta,k+k_p}), \tag{30}$$

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

We need some care in manipulating the Coulomb-induced terms, i.e., the last line. In order to do this, we reelaborated a technique used in the semiclassical context, which, once applied to four-wave mixing in quantum wells, allowed us to understand a wide range of observed phenomena.[14] In the following, we will show how, under this approximation, we 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. Written explicitly, it is

$$\frac{d}{dt}\hat{B}_{\pm\mathbf{k}}\bigg|_{\mathrm{Coul}}=-\frac{i}{\hbar}\sum_{\tilde{\sigma}\tilde{\mathbf{k}}}\hbar(\omega_{2(2\mathbf{k}_p)}-\omega_{1\tilde{\mathbf{k}}}-\omega_{1\mathbf{k}})\langle 1\tilde{\sigma}\tilde{\mathbf{k}}|\hat{B}_{\pm\mathbf{k}}|2(2\sigma_p2\mathbf{k}_p)\rangle\hat{X}_{1\tilde{\sigma}\tilde{\mathbf{k}},0}X_{0,2(2\sigma_p2\mathbf{k}_p)}-\frac{i}{\hbar}\hbar(\omega_{2(\mathbf{k}+\mathbf{k}_p)}-\omega_{1\mathbf{k}_p}-\omega_{1\mathbf{k}})$$
$$\times\langle 1\sigma_p\mathbf{k}_p|\hat{B}_{\pm\mathbf{k}}|2(\sigma_{\mathbf{k}+\mathbf{k}_p}\mathbf{k}+\mathbf{k}_p)\rangle X_{1\sigma_p\mathbf{k}_p,0}\hat{X}_{0,2[\sigma_{\mathbf{k}+\mathbf{k}_p}(\mathbf{k}+\mathbf{k}_p)]}. \tag{31}$$

Strictly speaking, from the point of view of the attaching strategy that we shall follow, the two terms need similar manipulations. Once the formal solution from the two-pair (biexciton) sector is plugged, we shall reformulate the exciton-photon source terms in $\hat{X}_{0;2}$, exploiting a dynamical identity that we shall write explicit in due course. The next steps comprise of an integration by parts, a summation over the biexciton sector, and some algebraic elementary manipulations of the interaction matrix elements, where we shall take into account that we are not considering situations where polarization mixing is present. As regard for the first line in Eq. (31), $X_{0,2(2\sigma_p2\mathbf{k}_p)}$ is a $\mathbb{C}$–number and there is no mathematical subtleties when readapting the procedure of Ref. 14 to the present context. As for the other term, which produces the shifts, we would exploit the formal biexciton solution as

$$\hat{X}_{0;2(k+k_p)}(t)=\int_{-\infty}^{t}dt'e^{-i\omega_{2(k+k_p)}(t-t')}\frac{i}{\hbar}\left(V_{k_p}\langle 2(k+k_p)|\hat{B}_{k_p}^{\dagger}|1k\rangle\hat{X}_{0,1k}Y_{0,1k_p}+V_k\langle 2(k+k_p)|\hat{B}_k^{\dagger}|1k_p\rangle X_{0,1k_p}\hat{Y}_{0,1k}\right), \tag{32}$$

where, for the sake of consistence, we are neglecting $\hat{X}_{0;2(k+k_p)}(-\infty)$ 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 arrives. Moreover, in the linear dynamics, we can exploit the following identity:

$$\frac{d}{dt}(\hat{X}_{0,1k}X_{0,1k_p}e^{-i(\omega_{1k}+\omega_{1k_p})(t-t')})=\left(i\frac{V_k}{\hbar}\hat{Y}_{0,1k}X_{0,1k_p}+i\frac{V_{k_p}}{\hbar}Y_{0,1k_p}\hat{X}_{0,1k}\right)e^{-i(\omega_{1k}+\omega_{1k_p})(t-t')}. \tag{33}$$

Then, we have

$$-\frac{i}{\hbar}\hbar(\omega_{2(\mathbf{k}+\mathbf{k}_p)}-\omega_{1\mathbf{k}_p}-\omega_{1\mathbf{k}})\langle 1\sigma_p\mathbf{k}_p|\hat{B}_{\pm\mathbf{k}}|2(\sigma_{\mathbf{k}+\mathbf{k}_p}\mathbf{k}+\mathbf{k}_p)\rangle X_{1\sigma_p\mathbf{k}_p,0}\hat{X}_{0,2[\sigma_{\mathbf{k}+\mathbf{k}_p}(\mathbf{k}+\mathbf{k}_p)]}=-\frac{i}{\hbar}\hbar X_{1\sigma_p\mathbf{k}_p,0}(t)\Bigg\{\Bigg[\langle 1\sigma_p\mathbf{k}_p|\hat{B}_{\pm\mathbf{k}}\Bigg(\frac{\hat{H}_e}{\hbar}-\omega_{1\mathbf{k}_p}$$
$$-\omega_{1\mathbf{k}}\Bigg)e^{-i(\hat{H}_e/\hbar)(t-t')}\hat{B}_k^{\dagger}|1\sigma_p\mathbf{k}_p\rangle\hat{X}_{0,1\pm\mathbf{k}}(t')\hat{X}_{0,1\sigma_p\mathbf{k}_p}(t')\Bigg]_{-\infty}^{t}-\int_{-\infty}^{t}dt'\hat{X}_{0,1\pm\mathbf{k}}(t')X_{0,1\sigma_p\mathbf{k}_p}(t')e^{-i(\omega_{1k}+\omega_{1k_p})(t-t')}$$
$$\times\frac{d}{dt}\Bigg[\langle 1\sigma_p\mathbf{k}_p|\hat{B}_{\pm\mathbf{k}}\Bigg(\frac{\hat{H}_e}{\hbar}-\omega_{1\mathbf{k}_p}-\omega_{1\mathbf{k}}\Bigg)e^{-i(\hat{H}_e/\hbar)(t-t')}\hat{B}_k^{\dagger}|1\sigma_p\mathbf{k}_p\rangle\Bigg]\Bigg\}=-\frac{i}{\hbar}X_{1\sigma_p\mathbf{k}_p,0}(t)$$
$$\times\Bigg\{\hbar\langle 0|\hat{D}_{\sigma_p\mathbf{k}_p,\pm\mathbf{k}}\hat{B}_{\pm\mathbf{k}}^{\dagger}\hat{B}_{\sigma_p\mathbf{k}_p}^{\dagger}|0\rangle\hat{X}_{0,1\pm\mathbf{k}}(t)X_{0,1\sigma_p\mathbf{k}_p}(t)-\int_{-\infty}^{t}dt'\hbar\langle 0|\hat{D}_{\sigma_p\mathbf{k}_p,\pm\mathbf{k}}(t-t')\hat{D}_{\sigma_p\mathbf{k}_p,\pm\mathbf{k}}^{\dagger}|0\rangle\hat{X}_{0,1\pm\mathbf{k}}(t')X_{0,1\sigma_p\mathbf{k}_p}(t')\Bigg\}. \tag{34}$$

Once similar manipulations for the remaining part in Eq. (31) are performed, by adding the phase-space-filling contribution and considering the case of input light beams activating only excitons with all the same circularly (e.g., σ^+) polarization, the final result reads

$$\frac{d}{dt}\hat{B}_{\pm\mathbf{k}} = -i\omega_{\mathbf{k}}\hat{B}_{\pm\mathbf{k}} + i\frac{V}{\hbar}\hat{a}_{\pm\mathbf{k}} - \frac{i}{\hbar}\frac{V}{n_{\mathrm{sat}}}(\hat{X}_{1\pm\mathbf{k}_i,0}X_{0,1\pm\mathbf{k}_p}Y_{0,1\pm\mathbf{k}_p} + X_{1\pm\mathbf{k}_p,0}X_{0,1\pm\mathbf{k}_p}\hat{Y}_{0,1\pm\mathbf{k}} + X_{1\pm\mathbf{k}_p,0}\hat{X}_{0,1\pm\mathbf{k}}Y_{0,1\pm\mathbf{k}_p}) - \frac{i}{\hbar}\hat{X}_{1\pm\mathbf{k}_i,0}(t)$$
$$\times\left\{V_{xx}X_{0,1\pm\mathbf{k}_p}(t)X_{0,1\pm\mathbf{k}_p}(t) - i\int_{-\infty}^{t}dt'F^{\pm\pm}(t-t')X_{0,1\pm\mathbf{k}_p}(t')X_{0,1\pm\mathbf{k}_p}(t')\right\} - 2\frac{i}{\hbar}X_{1\pm\mathbf{k}_p,0}(t)\left\{V_{xx}\hat{X}_{0,1\pm\mathbf{k}}(t)X_{0,1\pm\mathbf{k}_p}(t)\right.$$
$$\left. - i\int_{-\infty}^{t}dt'F^{\pm\pm}(t-t')\hat{X}_{0,1\pm\mathbf{k}}(t')X_{0,1\pm\mathbf{k}_p}(t')\right\}, \tag{35}$$

where $\mathbf{k}_i = 2\mathbf{k}_p - \mathbf{k}$, and

$$V_{xx} = \frac{\hbar}{2}\langle 0|\hat{D}_{\tilde{k},k}\hat{B}^{\dagger}_{k'}\hat{B}^{\dagger}_{k''}|0\rangle,$$

$$F^{k'',k'}_{\tilde{k},k}(t-t') = \frac{\hbar}{2}\langle 0|\hat{D}_{\tilde{k},k}(t-t')\hat{D}^{\dagger}_{k'',k'}|0\rangle,$$

$$\hat{D}_{\tilde{k},k} \doteq [\hat{B}_{\tilde{k}},[\hat{B}_k,\hat{H}_c]], \tag{36}$$

where a *force* operator $\hat{D}$ is defined.[14] In order to lighten the notation, we dropped the two spin indexes σ and $\tilde{\sigma}$ in the four-particle kernel function F defined in Eq. (36) for they are already univocally determined once the others are chosen (i.e., σ' and σ'') as soon as their selection rule ($\delta_{\sigma+\tilde{\sigma};\sigma'+\sigma''}$) is applied. In the specific case under analysis, we are considering cocircularly polarized waves and more explicit calculations of the nonlinear Coulomb term in Eq. (36) can be found in Refs. 31 and 32. The generalization to arbitrary polarization can be derived by translating the semiclassical derivation of Ref. 33 within this full quantum framework.

For the range of $\mathbf{k}$ space of interest, i.e., $|\mathbf{k}| \ll \frac{\pi}{a_x}$ (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., cocircularly polarized waves) conserve the polarizations, $F^{\pm\mp}(t-t')$ and $F^{\mp\pm}(t-t')$ (countercircular polarization) give rise to a mixing between the two circularly polarizations. The physical origin of the terms in Eq. (35) 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 and 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.[14]

The strong exciton-photon coupling does not modify the memory kernel because four-particle correlations do not directly couple to cavity photons. As pointed out clearly in Ref. 18, 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 with respect to that of excitons in bare QWs, thus producing a modification of the integral in Eq. (35). This way the exciton-photon coupling V_{nk} affects the exciton-exciton collisions that govern the polariton amplification process. Reference 18 considers the first (mean-field) and the second (four-particle correlation) terms in the particular case of cocircularly polarized waves, calling them without indices as V_{xx} and $F(t)$, respectively. In Fig. 1, of Ref. 18, they show $\mathcal{F}(\omega)$, as the Fourier transform of $F(t)$ plus the mean-field term V_{xx},

$$\mathcal{F}(\omega) = V_{xx} - i\int_{-\infty}^{\infty}dtF(t)e^{i\omega t}. \tag{37}$$

Its imaginary part is responsible for the frequency dependent excitation induced dephasing. It reflects the density of the states of two-exciton pair coherences. Toward the negative detuning region, the dispersive part $\mathrm{Re}(\mathcal{F})$ increases, whereas the absorptive part $\mathrm{Im}(\mathcal{F})$ 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 $V_{xx}n_{\mathrm{sat}} \simeq 11.39$ meV, which is clearly an upper bound for $\mathrm{Re}(\mathcal{F})$ 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[18] that both effects depends on the sum of the energies of the scattered polariton pairs.

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

$$\frac{d}{dt}\hat{a}_{\pm\mathbf{k}} = -i\omega^{c}_{\mathbf{k}}\hat{a}_{\pm\mathbf{k}} + i\frac{V}{\hbar}\hat{B}_{\pm\mathbf{k}} + t_c\frac{E_{\pm\mathbf{k}}}{\hbar},$$

$$\frac{d}{dt}\hat{B}_{\pm k} = -i\omega_{\mathbf{k}}\hat{B}_{\pm\mathbf{k}} + \hat{s}_{\pm\mathbf{k}} + i\frac{V}{\hbar}\hat{a}_{\pm\mathbf{k}} - \frac{i}{\hbar}R^{NL}_{\pm\mathbf{k}}, \tag{38}$$

where $R^{NL}_{\pm\mathbf{k}} = (R^{sat}_{\pm\mathbf{k}} + R^{xx}_{\pm\mathbf{k}})$,

$$R^{sat}_{\pm\mathbf{k}} = \frac{V}{n_{\mathrm{sat}}}B_{\pm\mathbf{k}_p}a_{\pm\mathbf{k}_p}\hat{B}^{\dagger}_{\pm\mathbf{k}_i},$$

$$R^{xx}_{\pm\mathbf{k}} = \hat{B}^{\dagger}_{\pm\mathbf{k}_i}(t)\left(V_{xx}B_{\pm\mathbf{k}_p}(t)B_{\pm\mathbf{k}_p}(t) - i\int_{-\infty}^{t}dt'F^{\pm\pm}(t-t')\right.$$
$$\left.B_{\pm\mathbf{k}_p}(t')B_{\pm\mathbf{k}_p}(t')\right). \tag{39}$$

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

$$\hat{s}_{\pm\mathbf{k}} = -i\left(\frac{V}{n_{\mathrm{sat}}}(B^*_{\pm\mathbf{k}_p}a_{\pm\mathbf{k}_p}\hat{B}_{\pm\mathbf{k}} + B^*_{\pm\mathbf{k}_p}B_{\pm\mathbf{k}_p}\hat{a}_{\pm\mathbf{k}}) + 2\frac{V_{xx}}{\hbar}B^*_{\pm\mathbf{k}_p}B_{\pm\mathbf{k}_p}\hat{B}_{\pm\mathbf{k}} - 2\frac{i}{\hbar}B^*_{\pm\mathbf{k}_p}(t)\int_{-\infty}^{t}dt' F^{\pm\pm}(t-t')\hat{B}_{\pm\mathbf{k}}(t')B_{\pm\mathbf{k}_p}(t')\right). \quad (40)$$

Equation (38) is the main result of this paper. They can be considered as 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 noninstantaneous four-particle correlation, to quantum optical effects. Starting from here, in the strong-coupling case, it might be useful to transform the description into a polariton basis. As a consistency check, as soon as we take the expectation values of Eq. (38), we obtain a result analogous to the corresponding equations describing the semiclassical (quantized electron system and classical light field) coherent $\chi^{(3)}$ response in a QW;[14] the main difference is that the (intracavity) light field here is regarded not as a driving external source but as a dynamical field.[18] This close correspondence for the dynamics of expectation values of the exciton operators is a consequence of the linearization of quantum fluctuations. Nevertheless, completely different results can be obtained for exciton or photon number expectation values or for higher-order correlation function.[10,25] Moreover, the present approach includes the light-field quantization and can thus be applied to the description of quantum optical phenomena. Even if formally similar to that of Ref. 14, they represent 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. 14, the electromagnetic field entered as a parameter only. Former analogous extensions have been obtained within a semiclassical model.[18,31,32] Once the expectations values are taken, Eq. (38) are 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., appendices), a restricted class of higher-order effects can be obtained from solving Eq. (38) 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.[14,18,21] Such multiple-scattering processes are expected to be very effective in cavity-embedded QW's due to multiple reflections at the Bragg mirrors.

V. PICTURE OF INTERACTING POLARITONS

This section is devoted to introduce and comment the picture of interacting polaritons within our quantum-DCTS framework. We shall consider strengths and weaknesses of this approach even in comparison with existing theories.[28,34] The former considers an effective model Hamiltonian describing excitons as interacting bosons; we will find that the resulting equations formally well reproduce the Hartree–Fock mean-field dynamics, enabling the author to catch the essential physical picture. Nevertheless, the resulting interaction terms due to phase space filling quantitatively differ from those obtained within our approach. It seems worth underlining that, thanks to the employed DCTS, we are left with the exact dynamical equations in the $\chi^{(3)}$ limit with correct matrix elements. From this point of view, our scheme results in a logically subsequent refinement of the previous theory. In Ref. 34, the problem of getting rid of exciton bosonization has been pursued; they calculated the corrections to bosonic commutation rules for excitons putting them explicitly into play throughout their calculations. Anyway, it is based on a Fermi golden rule description of the Coulomb induced interaction processes, which is unable to provide an accurate description of coherent dynamics of the optical response. As a consequence, our quantum-DCTS framework results in a rigorous and accurate tool for a theoretical description of quantum optics experiments in the strong-coupling regime, as we showed in Ref. 22, where the first application of the present theoretical framework has been presented.

Equation (38) can be written in compact form as (we drop the spin indices for simplicity)

$$\dot{\mathcal{B}}_{\mathbf{k}} = -i\mathbf{\Omega}^{\mathrm{xc}}_{\mathbf{k}}\mathcal{B}_{\mathbf{k}} + \mathcal{E}_{\mathbf{k}} - i\mathcal{R}^{NL}_{\mathbf{k}}, \quad (41)$$

where

$$\mathcal{B}_{\mathbf{k}} \equiv \begin{pmatrix}\hat{B}_{\mathbf{k}}\\ \hat{a}_{\mathbf{k}}\end{pmatrix}, \quad \mathbf{\Omega}^{\mathrm{xc}}_{\mathbf{k}} \equiv \begin{pmatrix}\omega^x_{\mathbf{k}} & -V\\ -V & \omega^c_{\mathbf{k}}\end{pmatrix},$$

$$\mathcal{E}_{\mathbf{k}} \equiv \begin{pmatrix}0\\ t_c E_{\mathbf{k}}\end{pmatrix}, \quad \text{and } \mathcal{R}^{NL}_{\mathbf{k}} \equiv \begin{pmatrix}\hat{s}_{\mathbf{k}} + \hat{R}^{NL}_{\mathbf{k}}\\ 0\end{pmatrix}.$$

Strong coupling occurs when the coupling rate V exceeds the decay rate of the exciton coherence and of the cavity field. In this regime, continuous exchanges of energy between excitons and photons become so intense that, before decaying, significantly alter the dynamics and hence the resulting resonances of the coupled system with respect to those of bare excitons and cavity photons. In this sense, cavity polaritons arise as the two-dimensional eigenstates of $\Omega^{\mathrm{xc}}_{\mathbf{k}}$. The coupling rate V determines the splitting ($\simeq 2$ V) between the two polariton energy bands. This nonperturbative dynamics including the interactions (induced by $\hat{R}^{NL}_{\mathbf{k}}$) between different polariton modes can be accurately described by Eq. (38). Nevertheless, there can be reasons to prefer a change of bases from excitons and photons to the eigenstates of the coupled system, namely, polaritons. An interesting one is that the resulting equations may provide a more intuitive description of nonlinear optical processes in terms of interacting polaritons. Moreover, equations describing the nonlinear interactions between polaritons become more similar to those describing parametric interactions between photons widely adopted in quantum optics. Another, more fundamental reason, is that the standard second-order Born–Markov approximation scheme, which is usually adopted to describe the interaction with environment, is strongly bases dependent, and

using the eigenstates of the closed system provides more accurate results. In order to diagonalize, we perform the unitary basis transformation

$$\mathcal{P}_{\mathbf{k}} = U_{\mathbf{k}} \mathcal{B}_{\mathbf{k}}, \tag{42}$$

where

$$\mathcal{P}_{\mathbf{k}} = \begin{pmatrix} \hat{P}_{1\mathbf{k}} \\ \hat{P}_{2\mathbf{k}} \end{pmatrix}$$

and

$$U_{\mathbf{k}} = \begin{pmatrix} X_{1\mathbf{k}} & C_{1\mathbf{k}} \\ X_{2\mathbf{k}} & C_{2\mathbf{k}} \end{pmatrix}. \tag{43}$$

In general, photon operators obey Bose statistics; on the contrary, the excitons do not posses a definite statistics (i.e., either bosonic or fermionic), but their behavior may be well approximated by a bosoniclike statistics in the limit of low excitation densities. Indeed,

$$[\hat{B}_n, \hat{B}^{\dagger}_{n'}] = \delta_{n',n} - \sum_{\mathbf{q}} \Phi^*_{n\mathbf{q}} \Phi_{n'\mathbf{q}} \sum_{N,\alpha,\beta} \left(\langle N\alpha | \hat{c}^{\dagger}_{\mathbf{q}} c_{\mathbf{q}} | N\beta \rangle + \langle N\alpha | \hat{d}^{\dagger}_{-\mathbf{q}} d_{-\mathbf{q}} | N\beta \rangle \right) |N\alpha\rangle\langle N\beta|. \tag{44}$$

Thus, within a DCTS line of reasoning,[14] the expectation values of these transition operators (i.e., $|N\alpha\rangle\langle N\beta|$) are at least of the second order in the incident light field, i.e., they are density-dependent contributions. Evidently, all these considerations affect polariton statistics as well; polariton is a linear combination of intracavity photons and excitons. As a consequence, even if polariton operators have no definite statistics, in the limit of low excitation intensites, they obey approximately bosoniclike commutation rules.

Diagonalizing $\Omega^{\mathrm{xc}}_{\mathbf{k}}$,

$$U_{\mathbf{k}} \mathbf{\Omega}^{\mathrm{xc}}_{\mathbf{k}} = \tilde{\Omega}_{\mathbf{k}} U_{\mathbf{k}}, \tag{45}$$

where

$$\tilde{\Omega}_{\mathbf{k}} = \begin{pmatrix} \omega_{1\mathbf{k}} & 0 \\ 0 & \omega_{2\mathbf{k}} \end{pmatrix}.$$

Here, $\omega_{1,2}$ are the eigenenergy (as a function of $\mathbf{k}$) of the lower (1) and upper (2) polariton states. With Hopfield coefficients,[22]

$$X_{1\mathbf{k}} = \frac{1}{\sqrt{1 + \left(\frac{V}{\omega_{1\mathbf{k}} - \omega^c_{\mathbf{k}}} \right)^2}}, \quad C_{1\mathbf{k}} = \frac{1}{\sqrt{1 + \left(\frac{\omega_{1\mathbf{k}} - \omega^c_{\mathbf{k}}}{V} \right)^2}}. \tag{46}$$

By introducing this transformation into Eq. (41), one obtains

$$\dot{\mathcal{P}}_{\mathbf{k}} = -i \tilde{\Omega}_{\mathbf{k}} \mathcal{P}_{\mathbf{k}} + \tilde{\mathcal{E}}_{\mathbf{k}} - i \tilde{\mathcal{R}}^{NL}_{\mathbf{k}}, \tag{47}$$

where $\tilde{\mathcal{R}}^{NL} = U \mathcal{R}^{NL}$, which in explicit form reads

$$\dot{\hat{P}}_{1\mathbf{k}} = -i\omega_{1\mathbf{k}} \hat{P}_{1\mathbf{k}} - i\tilde{s}_{1\mathbf{k}} + \tilde{E}_{1\mathbf{k}} - i\tilde{R}^{NL}_{1\mathbf{k}}, \tag{48a}$$

$$\dot{\hat{P}}_{2\mathbf{k}} = -i\omega_{2\mathbf{k}} \hat{P}_{2\mathbf{k}} - i\tilde{s}_{2\mathbf{k}} + \tilde{E}_{2\mathbf{k}} - i\tilde{R}^{NL}_{2\mathbf{k}}, \tag{48b}$$

where $\tilde{E}_{m\mathbf{k}} = t_c C_{m\mathbf{k}} E_{\mathbf{k}}$ and $\tilde{R}^{NL}_{m\mathbf{k}} = X_{m\mathbf{k}} \hat{R}^{NL}_{\mathbf{k}}$ $(m = 1, 2)$. Such a diagonalization is the necessary step when the eigenstates of the polariton system are to be used as the starting states perturbed by the interaction with the environment degrees of freedom. The nonlinear interaction written in terms of polariton operators reads

$$\hat{R}^{NL}_{\mathbf{k}} = \sum_{i,j,l} \hat{P}^{\dagger}_{i\mathbf{k}_i}(t) \int_{-\infty}^{t} g^{ijl}_{m\mathbf{k}}(t,t') P_{j\mathbf{k}_p}(t') P_{l\mathbf{k}_p}(t') dt', \tag{49}$$

where

$$g^{ijl}_{m\mathbf{k}}(t,t') = \frac{1}{N_{eff}} \left[\frac{V}{n_{sat}} C^*_{j,\mathbf{k}_p} \delta(t-t') + (V_{xx}\delta(t-t') - iF(t-t')) X^*_{j,\mathbf{k}_p} \right] X^*_{l,\mathbf{k}_p} X_{i,\mathbf{k}_i}. \tag{50}$$

The shift $\hat{s}_{\mathbf{k}}(t)$ is transformed into

$$\tilde{s}_{m\mathbf{k}}(t) = \sum_{i,j,l} P^*_{i\mathbf{k}_p}(t) \int_{-\infty}^{t} (h^{ijl}_{m\mathbf{k}} \delta(t-t') - 2iF(t-t')) P_{j\mathbf{k}_p}(t') \hat{P}_{l\mathbf{k}}(t') dt' \tag{51}$$

and

$$h^{ijl}_{m\mathbf{k}} = \frac{1}{N_{eff}} X_{m\mathbf{k}} \left[\frac{V}{n_{sat}} X_{i\mathbf{k}_p} (C^*_{j,\mathbf{k}_p} X^*_{l\mathbf{k}} + X^*_{j\mathbf{k}_p} C^*_{l\mathbf{k}}) + 2V_{xx} X_{i\mathbf{k}_p} X^*_{j\mathbf{k}_p} X^*_{l\mathbf{k}} \right]. \tag{52}$$

Equations (48a) and (48b) describe the coherent dynamics of a system of interacting cavity polaritons. The nonlinear term drives the mixing between polariton modes with different in-plane wave vectors and possibly belonging to different branches.

Analogous equations can be obtained starting from an effective Hamiltonian describing excitons as interacting bosons.[28] 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 obtained within the present approach not based on an effective Hamiltonian. Indeed, Eq. (38) has nonlinear terms of the same structure of Ref. 28 [see Eqs. (43) and (44) even if they are already written in the polariton basis] but display two main differences originating from the different starting points. Our equations, obtained from the DCTS, includes the noninstantaneous four-particle correlation determining a correction to the mean-field Coulomb interaction and a frequency-dependent excitation-induced dephasing. Moreover, whereas the mean-field Coulomb interaction coincides in the two approaches, the interaction term originating from phase space filling differs.

In particular (for $N_{eff} = 1$, considering only lower polaritons, and for real Hopfield coefficients), we obtain

$$R_{\mathbf{k}}^{NL}|_{psf} = \frac{V}{n_{sat}} \sum_{\mathbf{k}',\mathbf{q}} C_{\mathbf{k}'-\mathbf{q}} X_{\mathbf{k}+\mathbf{q}} X_{\mathbf{k}'} P_{\mathbf{k}'}^{*} P_{\mathbf{k}+\mathbf{q}} P_{\mathbf{k}'-\mathbf{q}}. \quad (53)$$

The corresponding term in Ref. 28 can be written as

$$R_{\mathbf{k}}^{NL}|_{psf} = 2\frac{V}{n_{sat}} \sum_{\mathbf{k}',\mathbf{q}} C_{\mathbf{k}'-\mathbf{q}} X_{\mathbf{k}+\mathbf{q}} X_{\mathbf{k}'} P_{\mathbf{k}'}^{*} P_{\mathbf{k}+\mathbf{q}} P_{\mathbf{k}'-\mathbf{q}} + \frac{V}{n_{sat}} \sum_{\mathbf{k}',\mathbf{q}} C_{\mathbf{k}'} X_{\mathbf{k}+\mathbf{q}} X_{\mathbf{k}'-\mathbf{q}} P_{\mathbf{k}'}^{*} P_{\mathbf{k}+\mathbf{q}} P_{\mathbf{k}'-\mathbf{q}}, \quad (54)$$

which contains additional terms providing an interaction strength due to phase-space filling larger of about a factor 3 and displaying a different k dependence. We believe that the difference is maily due to the adopted bosonization procedure. According to that procedure, the exciton operator (determining the resonant polarization) $\hat{B}$ is expanded in terms of Bose operators $\hat{B}_B$ up to the first two terms. Schematically, $\hat{B} \rightarrow \hat{B}_B + \hat{B}_B^{\dagger}\hat{B}_B\hat{B}_B + \cdots$. Then, the equation of motion for $\langle \hat{B}_B \rangle$ is obtained. The discrepancy may arise from the fact that $\hat{B}_B + \hat{B}_B^{\dagger}\hat{B}_B\hat{B}_B$ and not $\hat{B}_B$ should be regarded as the proper polarization operator. It is worth noticing that more rigourously calculated nonlinear coupling coefficients will more accurately describe parametric dynamics, as evidenced by the good quantitative agreement with experimental data, which our numerical results show in Ref. 32, where we numerically tested our framework. 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, in the strong-coupling regime, will be presented in another paper of ours,[22] which extends the Heisenberg-Langevin approach already adopted for studying the linear optical response of QW excitons,[35] to arbitrary environment interaction Hamiltonians.

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 noninstantaneous 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.[22]

APPENDIX A: THE EQUATION OF MOTION AT ANY ORDER

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

$$\begin{aligned}
\frac{d}{dt}(\hat{X}_{N\alpha;M\beta}\hat{Y}_{n\lambda;m\mu}) = & -i\left(\omega_{M\beta} - \omega_{N\alpha} + \sum_{i=1}^{m}\omega_{k_i}^{c} - \sum_{j=1}^{n}\omega_{k_j}^{c}\right)(\hat{X}_{N\alpha;M\beta}\hat{Y}_{n\lambda;m\mu}) \\
& + \hat{X}_{N\alpha;M\beta}\left(\delta_{m,1}t_c\frac{E_\mu}{\hbar}\hat{Y}_{n\lambda;0} + \delta_{n,1}t_c\frac{E_\lambda^{*}}{\hbar}\hat{Y}_{0;m\mu}\right) - \hat{X}_{N\alpha;M\beta}\sum_k t_c\left(\delta_{m,0}\frac{E_k^{*}}{\hbar}\hat{Y}_{n\lambda;1k} + \delta_{n,0}\frac{E_k}{\hbar}\hat{Y}_{1k;m\mu}\right) \\
& + \sum_{k\nu} t_c\hat{X}_{N\alpha;M\beta}\Big[\Theta(m-2)\langle m\mu|\hat{a}_k^{\dagger}|(m-1)\nu\rangle\frac{E_k}{\hbar}\hat{Y}_{n\lambda;(m-1)\nu} \\
& + \Theta(n-2)\langle(n-1)\nu|\hat{a}_k|n\lambda\rangle\frac{E_k^{*}}{\hbar}\hat{Y}_{(n-1)\nu;m\mu} \\
& - \Theta(m-1)\langle m\mu|\hat{a}_k|(m+1)\nu\rangle\frac{E_k^{*}}{\hbar}\hat{Y}_{n\lambda;(m+1)\nu} \\
& - \Theta(n-1)\langle(n+1)\nu|\hat{a}_k^{\dagger}|n\lambda\rangle\frac{E_k}{\hbar}\hat{Y}_{(n+1)\nu;m\mu}\Big] \\
& + \frac{i}{\hbar}\delta_{M,0}\delta_{\beta,0}\delta_{m,1}\sum_{\bar{n}} V_{\bar{n}\mu}^{*}\hat{X}_{N\alpha;1\bar{n}\mu}\hat{Y}_{n\lambda;0} - \frac{i}{\hbar}\delta_{N,0}\delta_{\alpha,0}\delta_{n,1}\sum_{\bar{n}} V_{\bar{n}\lambda}\hat{X}_{1\bar{n}\lambda;M\beta}\hat{Y}_{0;m\mu} \\
& - \frac{i}{\hbar}\delta_{N,1}\delta_{n,0}\delta_{\lambda,0}V_\alpha^{*}\hat{X}_{0;M\beta}\hat{Y}_{1k_\alpha;m\mu} + \frac{i}{\hbar}\delta_{M,1}\delta_{m,0}\delta_{\mu,0}V_\beta\hat{X}_{N\alpha;0}\hat{Y}_{n\lambda;1k_\beta}
\end{aligned}$$

$$
\begin{aligned}
&+\frac{i}{\hbar}\delta_{m,1}\Theta(M-1)\sum_{\bar{n}\delta}V^{*}_{\bar{n}\mu}\langle M\beta|\hat{B}_{\bar{n}\mu}|(M+1)\delta\rangle\hat{X}_{N\alpha;(M+1)\delta}\hat{Y}_{n\lambda,0}\\
&-\frac{i}{\hbar}\delta_{n,1}\Theta(N-1)\sum_{\bar{n}\eta}V_{\bar{n}\lambda}\langle(N+1)\eta|\hat{B}^{\dagger}_{\bar{n}\lambda}|N\alpha\rangle\hat{X}_{(N+1)\eta;M\beta}\hat{Y}_{0;m\mu}\\
&-\frac{i}{\hbar}\delta_{n,0}\delta_{\lambda,0}\Theta(N-2)\sum_{\bar{n}k\eta}V^{*}_{\bar{n}k}\langle(N-1)\eta|\hat{B}_{\bar{n}k}|N\alpha\rangle\hat{X}_{(N-1)\eta;M\beta}\hat{Y}_{1k;m\mu}\\
&+\frac{i}{\hbar}\delta_{m,0}\delta_{\mu,0}\Theta(M-2)\sum_{\bar{n}k\delta}V_{\bar{n}k}\langle M\beta|\hat{B}^{\dagger}_{\bar{n}k}|(M-1)\delta\rangle\hat{X}_{N\alpha;(M-1)\delta}\hat{Y}_{n\lambda;1k}\\
&+\frac{i}{\hbar}\delta_{M,0}\delta_{\beta,0}\Theta(m-2)\sum_{\bar{n}k\nu}V^{*}_{\bar{n}k}\langle m\mu|\hat{a}^{\dagger}_{k}|(m-1)\nu\rangle\hat{X}_{N\alpha;1\bar{n}k}\hat{Y}_{n\lambda;(m-1)\nu}\\
&-\frac{i}{\hbar}\delta_{N,0}\delta_{\alpha,0}\Theta(n-2)\sum_{\bar{n}k\gamma}V_{\bar{n}k}\langle(n-1)\gamma|\hat{a}_{k}|n\lambda\rangle\hat{X}_{1\bar{n}k;M\beta}\hat{Y}_{(n-1)\gamma;m\mu}\\
&-\frac{i}{\hbar}\delta_{N,1}\Theta(n-1)V^{*}_{\alpha}\sum_{\gamma}\langle(n+1)\gamma|\hat{a}^{\dagger}_{k_{\alpha}}|n\lambda\rangle\hat{X}_{0;M\beta}\hat{Y}_{(n+1)\gamma;m\mu}\\
&+\frac{i}{\hbar}\delta_{M,1}\Theta(m-1)V_{\beta}\sum_{\nu}\langle m\mu|\hat{a}_{k_{\beta}}|(m+1)\nu\rangle\hat{X}_{N\alpha;0}\hat{Y}_{n\lambda;(m+1)\nu}\\
&+\frac{i}{\hbar}\sum_{\bar{n}k}\sum_{\nu\delta}\Big[V^{*}_{\bar{n}k}\Big(\Theta(M-1)\Theta(m-2)\langle M\beta|\hat{B}_{\bar{n}k}|(M+1)\delta\rangle\langle m\mu|\hat{a}^{\dagger}_{k}|(m-1)\nu\rangle\hat{X}_{N\alpha;(M+1)\delta}\hat{Y}_{n\lambda;(m-1)\nu}\\
&-\Theta(N-2)\Theta(n-1)\langle(N-1)\delta|\hat{B}_{\bar{n}k}|N\alpha\rangle\langle(n+1)\nu|\hat{a}^{\dagger}_{k}|n\lambda\rangle\hat{X}_{(N-1)\delta;M\beta}\hat{Y}_{(n+1)\nu;m\mu}\Big)\\
&-V_{\bar{n}k}\Big(\Theta(N-1)\Theta(n-2)\langle(N+1)\delta|\hat{B}^{\dagger}_{\bar{n}k}|N\alpha\rangle\langle(n-1)\nu|\hat{a}_{k}|n\lambda\rangle\hat{X}_{(N+1)\delta;M\beta}\hat{Y}_{(n-1)\nu;m\mu}\\
&-\Theta(M-2)\Theta(m-1)\langle M\beta|\hat{B}^{\dagger}_{\bar{n}k}|(M-1)\delta\rangle\langle m\mu|\hat{a}_{k}|(m+1)\nu\rangle\hat{X}_{N\alpha;(M-1)\delta}\hat{Y}_{n\lambda;(m+1)\nu}\Big)\Big]. \qquad \text{(A1)}
\end{aligned}
$$

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

APPENDIX B: N *eh* PAIR AGGREGATES

We start from the usual model for the electronic Hamiltonian of a direct two-band semiconductor.[12,23] It is obtained from the many-body Hamiltonian of the interacting electron system in a lattice, explicitly keeping only those terms in the Coulomb interaction preserving the number of electrons in a given band and can be expressed as

$$\hat{H}_e = \hat{H}_0 + \hat{V}_{\text{Coul}}. \qquad \text{(B1)}$$

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

$$\hat{H}_0 = \sum_k E_{c,k}\hat{c}^{\dagger}_k\hat{c}_k + \sum_k E_{h,k}\hat{d}^{\dagger}_k\hat{d}_k, \qquad \text{(B2)}$$

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,

$$
\begin{aligned}
\hat{V}_{\text{Coul}} = &\frac{1}{2}\sum_{\mathbf{q}\neq 0}V_q\sum_{\sigma,\mathbf{k},\sigma',\mathbf{k}'}\hat{c}^{\dagger}_{\sigma,\mathbf{k}+\mathbf{q}}\hat{c}^{\dagger}_{\sigma',\mathbf{k}'-\mathbf{q}}\hat{c}_{\sigma',\mathbf{k}'}\hat{c}_{\sigma,\mathbf{k}}\\
&+\frac{1}{2}\sum_{\mathbf{q}\neq 0}V_q\sum_{\sigma,\mathbf{k},\sigma',\mathbf{k}'}\hat{d}^{\dagger}_{\sigma,\mathbf{k}+\mathbf{q}}\hat{d}^{\dagger}_{\sigma',\mathbf{k}'-\mathbf{q}}\hat{d}_{\sigma',\mathbf{k}'}\hat{d}_{\sigma,\mathbf{k}}\\
&-\sum_{\mathbf{q}\neq 0}V_q\sum_{\sigma,\mathbf{k},\sigma',\mathbf{k}'}\hat{c}^{\dagger}_{\sigma,\mathbf{k}+\mathbf{q}}\hat{d}^{\dagger}_{\sigma',\mathbf{k}'-\mathbf{q}}\hat{d}_{\sigma',\mathbf{k}'}c_{\sigma,\mathbf{k}}. \qquad \text{(B3)}
\end{aligned}
$$

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

$$\hat{H}_e|\Psi\rangle = (\hat{H}_0 + \hat{V}_{\text{Coul}})|\Psi\rangle = E|\Psi\rangle, \qquad \text{(B4)}$$

with $|\Psi\rangle$ as 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., $\hat{N}_e = \Sigma_k \hat{c}^{\dagger}_k\hat{c}_k$ and $\hat{N}_h = \Sigma_k \hat{d}^{\dagger}_k\hat{d}_k$.

Therefore, the state $|\Psi\rangle$ may build up corresponding on a given number of electrons and of holes. Moreover, because we shall consider the case of intrinsic semiconductors materials where $N_e=N_h \doteq N$, the good quantum number for the Schrödinger [Eq. (B4)] is explicitly the total number of electron-hole pairs N,

$$\hat{H}_e|N\alpha\rangle = E_{N\alpha}|N\alpha\rangle, \tag{B5}$$

where α is the whole set of proper quantum numbers needed to univocally specify the many-body state.

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

$$\hat{H}_0|Na\rangle = \epsilon_{Na}|Na\rangle, \tag{B6}$$

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

$$|N,a\rangle = \otimes_{n=1}^{N} \hat{c}^{\dagger}_{j_{en}} \hat{d}^{\dagger}_{j_{hn}}|0,0\rangle \quad \text{and} \quad \epsilon_{Na} = \sum_{n=1}^{N} (\epsilon_{j_{en}} + \epsilon_{j_{hn}}). \tag{B7}$$

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

$$|N\alpha\rangle = \sum_{a} U_a^{N\alpha}|Na\rangle. \tag{B8}$$

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, i.e., the solution of the corresponding secular equation. Indeed, the eigenvalue problem [Eq. (B5)] is transformed into

$$\sum_{a'} (\langle Na|\hat{H}_e|Na'\rangle - E_{N\alpha}\delta_{a,a'})U_{a'}^{N\alpha} = 0. \tag{B9}$$

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 detail 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 three-dimensional (3D) lattice]. Using the general mapping,[36] $\Sigma_{r_i} \leftrightarrow (1/v_0)\int d^3r$, $\delta(\mathbf{r}-\mathbf{r}') = (\delta_{\mathbf{r}_i,\mathbf{r}_j}/v_0)$, and $\hat{a}^{\dagger}_{c/v,\mathbf{r}_i} = [\hat{a}^{\dagger}_{c/v}(\mathbf{r})/\sqrt{v_0}]$ (creation operator of the conduction- or valence-band electron), where 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

$$\sum_{\mathbf{r}'_e,\mathbf{r}'_h} (\langle \mathbf{r}_e,\mathbf{r}_h|\hat{H}_e|\mathbf{r}'_e,\mathbf{r}'_h\rangle - E_{n\sigma\mathbf{k}}\delta_{\mathbf{r}_e\mathbf{r}_h,\mathbf{r}'_e\mathbf{r}'_h})U_a^{\alpha}(\mathbf{r}'_e,\mathbf{r}'_h) = 0, \tag{B10}$$

with

$$\langle \mathbf{r}_e,\mathbf{r}_h|\hat{H}_e|\mathbf{r}'_e,\mathbf{r}'_h\rangle = \left(-\frac{\hbar^2}{2m_e}\nabla^2_{r_e} - \frac{\hbar^2}{2m_h}\nabla^2_{r_h} - \frac{e^2}{\varepsilon_r|\mathbf{r}_e-\mathbf{r}_h|} + V(\mathbf{r}_e,\mathbf{r}_h)\right)\delta_{\mathbf{r}_e\mathbf{r}_h,\mathbf{r}'_e\mathbf{r}'_h}. \tag{B11}$$

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 is much larger than the Coulomb interaction between particles. As a consequence, at least at low temperatures, particles are confined at the lowest quantization level and the (possible) distortion 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 two-dimensional (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[37]). In addition, in QWs, light and heavy holes in valence band are split off in energy. By 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 as

$$\left(-\frac{\hbar^2}{2m_e}\frac{d^2}{dz_e^2} + V^e(z_e) - \frac{\hbar^2}{2m_h}\frac{d^2}{dz_h^2} + V^h(z_h)\right)U^{\alpha}(\mathbf{r}_e,\mathbf{r}_h) = E^z U^{\alpha}(\mathbf{r}_e,\mathbf{r}_h). \tag{B12}$$

It means we are solving our secular equation with solutions built up as linear combination of $F^{\alpha}_{n_c,n_v,a}(\mathbf{r}^{\parallel}_e,\mathbf{r}^{\parallel}_h)c_{n_c}(z_e)v_{n_v}(z_h)$, with $\mathbf{r}=(\mathbf{r}^{\parallel},z)$. 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 indices could still arise in due course.

Projecting Eq. (B10) on these confined states, we end up with an effective Schrödinger equation in the plane,

$$\left(-\frac{\hbar^2}{2m_e}\nabla^2_{\mathbf{r}^{\parallel}_e} - \frac{\hbar^2}{2m_h}\nabla^2_{\mathbf{r}^{\parallel}_h} - U_{n_c,n_v;n'_c,n'_v}(|\mathbf{r}^{\parallel}_e - \mathbf{r}^{\parallel}_h|)\right)F^{\alpha}_{n_c,n_v,a}(\mathbf{r}^{\parallel}_e,\mathbf{r}^{\parallel}_h) = (E_{\alpha} - E^z_{n_c} - E^z_{n_v})F^{\alpha}_{n_c,n_v,a}(\mathbf{r}^{\parallel}_e,\mathbf{r}^{\parallel}_h), \tag{B13}$$

with

$$U_{n_c,n_v;n_c',n_v'}(|\mathbf{r}_\mathbf{e}^\parallel - \mathbf{r}_\mathbf{h}^\parallel|) = \int dz_e \int dz_h \frac{e^2}{\varepsilon_r \sqrt{|\mathbf{r}_e^\parallel - \mathbf{r}_h^\parallel|^2 + (z_e - z_h)^2}} c_{n_c}(z_e) c_{n_c'}(z_e) v_{n_v}(z_h) v_{n_v'}(z_h). \tag{B14}$$

For what already stated, we shall consider only the lowest confined subband levels, then the resulting effective in-plane secular equation becomes

$$\left(-\frac{\hbar^2}{2m_e}\nabla^2_{\mathbf{r}_\mathbf{e}^\parallel} - \frac{\hbar^2}{2m_h}\nabla^2_{\mathbf{r}_\mathbf{h}^\parallel} - \int dz_e \int dz_h \frac{e^2}{\varepsilon_r \sqrt{|\mathbf{r}_e^\parallel - \mathbf{r}_h^\parallel|^2 + (z_e - z_h)^2}} |c(z_e)|^2 |v(z_h)|^2 \right) F_a^\alpha(\mathbf{r}_e^\parallel, \mathbf{r}_h^\parallel) = (E_\alpha - E_c^z - E_v^z) F_a^\alpha(\mathbf{r}_e^\parallel, \mathbf{r}_h^\parallel), \tag{B15}$$

with the product exciton envelope function $U^\alpha(\mathbf{r}_e, \mathbf{r}_h) = F_a^\alpha(\mathbf{r}_e^\parallel, \mathbf{r}_h^\parallel) c(z_e) v(z_h)$. Equation (B15) is solvable by separation of variables once we employ a coordinate transformation into center of mass (c.m.) $\mathbf{R} = (m_e \mathbf{r}_e^\parallel + m_h \mathbf{r}_h^\parallel)/(m_e + m_h)$ and relative $\boldsymbol{\rho} = (\mathbf{r}_e^\parallel - \mathbf{r}_h^\parallel)$ exciton coordinates. It reads

$$\left(-\frac{\hbar^2}{2M}\nabla^2_{\mathbf{R}} - \frac{\hbar^2}{2\mu}\nabla^2_{\boldsymbol{\rho}} - U(\boldsymbol{\rho}) \right) F_a^\alpha(\mathbf{R}, \boldsymbol{\rho}) = E F_a^\alpha(\mathbf{R}, \boldsymbol{\rho}), \tag{B16}$$

with a solution we can arrange as $F_a^\alpha(\mathbf{R}, \boldsymbol{\rho}) = \frac{e^{i\mathbf{K}\cdot\mathbf{R}}}{\sqrt{A}} \phi_a^\alpha(\boldsymbol{\rho})$ the latter being the solution of the relative hydrogenlike 2D problem.

Eventually, in real-space representation, we have our exciton wave function with total in-plane c.m. wave vector $\mathbf{K}$ (A is the in-plane quantization surface in the free directions) which reads

$$|n\sigma\mathbf{K}\rangle = \frac{v_0}{\sqrt{A}} \sum_{\mathbf{r_e}, r_\mathbf{h}} e^{i\mathbf{K}\cdot\mathbf{R}} \phi^\mathbf{K}_{n\sigma}(\boldsymbol{\rho}) c(z_e) v(z_h) a^\dagger_{c,\mathbf{r}_e} a_{v,\mathbf{r}_h} |0\rangle, \tag{B17}$$

where $\hat{a}^\dagger_{c/v,\mathbf{r}}(\hat{a}_{c/v,\mathbf{r}})$ is the creation (annihilation) operator of the conduction or valence-band electron in the Wannier representation and $\mathbf{r}_{e/h} = (\mathbf{r}^\parallel_{e/h}, z_{e/h})$ are to be considered coordinates of the direct lattice; $|0\rangle$ 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 ($Nv_0 = AL$ is the number of unit cells and L is the quantization dimension along the confined direction, $\nu = c, v$),

one obtains:

$$|n\sigma\mathbf{K}\rangle = \sum_{\substack{\mathbf{k},\mathbf{k}' \\ k_z,k_z'}} \delta_{\mathbf{K},\mathbf{k}-\mathbf{k}'} \left(\frac{1}{\sqrt{AL}} \int d^2\rho \int dz_e \int dz_h \phi^\mathbf{K}_{n\sigma}(\boldsymbol{\rho}) c(z_e) v(z_h) e^{-i\rho\cdot(\eta_h\mathbf{k} + \eta_e\mathbf{k}')} e^{-ik_z z_e} e^{+ik_z' z_h} \right) a^\dagger_{c,(\mathbf{k},k_z)} a_{v,(\mathbf{k}',k_z')} |0\rangle. \tag{B19}$$

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

$$\begin{cases} \mathbf{K} = \mathbf{k} - \mathbf{k}' \\ \mathbf{k}_r = \eta_h \mathbf{k} + \eta_e \mathbf{k}' \end{cases} \Rightarrow \begin{cases} \mathbf{k} = \mathbf{k}_r + \eta_e \mathbf{K} \\ \mathbf{k}' = \mathbf{k}_r - \eta_h \mathbf{K} \end{cases}. \tag{B20}$$

It becomes

$$|n\sigma\mathbf{K}\rangle = \sum_{\mathbf{K},\mathbf{k}_r} \delta_{\mathbf{K},\mathbf{k}-\mathbf{k}'} \sum_{k_z,k_z'} \left(\frac{1}{\sqrt{A}} \int d^2\rho \phi^\mathbf{K}_{n\sigma}(\boldsymbol{\rho}) e^{-i\rho\cdot\mathbf{k}_r} \right) \left(\frac{1}{\sqrt{L}} \int dz_e c(z_e) e^{-ik_z z_e} \right) \left(\frac{1}{\sqrt{L}} \int dz_h v(z_h) e^{+ik_z' z_h} \right) a^\dagger_{c,(\mathbf{k}_r + \eta_e\mathbf{K}, k_z)} a_{v,(\mathbf{k}_r - \eta_h\mathbf{K}, k_z')} |0\rangle. \tag{B21}$$

Thus,

$$|n\sigma\mathbf{K}\rangle = \sum_{\mathbf{k}_r} \sum_{k_z,k_z'} \Phi^\mathbf{K}_{n\sigma,\mathbf{k}_r} u^c_{k_z} u^{v*}_{k_z'} a^\dagger_{c,(\mathbf{k}_r + \eta_e\mathbf{K}, k_z)} a_{v,(\mathbf{k}_r - \eta_h\mathbf{K}, k_z')} |0\rangle, \tag{B22}$$

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

$$|n\sigma\mathbf{K}\rangle = \sum_{\mathbf{k}_r} \sum_{k_z,k_z'} \Phi^\mathbf{K}_{n\sigma,\mathbf{k}_r} u^e_{k_z} u^h_{k_z'} c^\dagger_{(\mathbf{k}_r + \eta_e\mathbf{K}, k_z)} d^\dagger_{(-\mathbf{k}_r + \eta_h\mathbf{K}, -k_z')} |0\rangle, \tag{B23}$$

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

*stefano.portolan@epfl.ch

[1] L. Esaki and R. Tsu, IBM J. Res. Dev. **14**, 61 (1970); L. Esaki and L. L. Chang, Phys. Rev. Lett. **33**, 495 (1974); R. Dingle, W. Wiegmann, and C. H. Henry, *ibid.* **33**, 827 (1974).

[2] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, 2000).

[3] C. Weisbuch, M. Nishioka, A. Ishikawa, and Y. Arakawa, Phys. Rev. Lett. **69**, 3314 (1992); R. Houdré, C. Weisbuch, R. P. Stanley, U. Oesterle, and M. Ilegems, Phys. Rev. Lett. **85**, 2793 (2000).

[4] R. M. Stevenson, V. N. Astratov, M. S. Skolnick, D. M. Emam-Ismail, A. I. Tartakovskii, P. G. Savvidis, J. J. Baumberg, and J. S. Roberts, Phys. Rev. Lett. **85**, 3680 (2000).

[5] J. Erland, V. Mizeikis, W. Langbein, J. R. Jensen, and J. M. Hvam, Phys. Rev. Lett. **86**, 5791 (2001).

[6] W. Langbein, Phys. Rev. B **70**, 205301 (2004).

[7] P. Schwendimann, C. Ciuti, and A. Quattropani, Phys. Rev. B **68**, 165324 (2003).

[8] S. Savasta, O. Di Stefano, V. Savona, and W. Langbein, Phys. Rev. Lett. **94**, 246401 (2005).

[9] S. Kundermann, M. Saba, C. Ciuti, T. Guillet, U. Oesterle, J. L. Staehli, and B. Deveaud, Phys. Rev. Lett. **91**, 107402 (2003).

[10] S. Savasta, G. Martino, and R. Girlanda, Solid State Commun. **111**, 495 (1999).

[11] J. Ph. Karr, A. Baas, R. Houdré, and E. Giacobino, Phys. Rev. A **69**, 031802(R) (2004).

[12] V. M. Axt and T. Kuhn, Rep. Prog. Phys. **67**, 433 (2004).

[13] K. Edamatsu, G. Oohata, R. Shimizu, and T. Itoh, Nature (London) **431**, 167 (2004).

[14] Th. Österich, K. Schönhammer, and L. J. Sham, Phys. Rev. Lett. **74**, 4698 (1995); Phys. Rev. B **58**, 12920 (1998).

[15] B. Krummheuer, V. M. Axt, T. Kuhn, I. D'Amico, and F. Rossi, Phys. Rev. B **71**, 235329 (2005).

[16] V. M. Axt and A. Stahl, Z. Phys. B: Condens. Matter **93**, 195 (1994); K. Victor, V. M. Axt, and A. Stahl, Phys. Rev. B **51**, 14164 (1995).

[17] S. Savasta and R. Girlanda, Phys. Rev. Lett. **77**, 4736 (1996).

[18] S. Savasta, O. Di Stefano, and R. Girlanda, Phys. Rev. Lett. **90**, 096403 (2003).

[19] See, e.g., D. F. Walls and G. J. Milburn, *Quantum Optics* (Springer-Verlag, Berlin, 1994).

[20] S. Savasta, O. Di Stefano, and R. Girlanda, Phys. Rev. B **64**, 073306 (2001).

[21] M. Buck, L. Wischmeier, S. Schumacher, G. Czycholl, F. Jahnke, T. Voss, I. Rückmann, and J. Gutowski, Eur. Phys. J. B **42**, 175 (2004).

[22] S. Portolan, S. Savasta, O. Di Stefano, F. Rossi, and R. Girlanda, Phys. Rev. B **77**, 035433 (2008).

[23] H. Haugh and S. W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors*, 2nd ed. (World Scientific, Singapore, 1993).

[24] K. V. Kavokin, I. A. Shelykh, A. V. Kavokin, G. Malpuech, and P. Bigenwald, Phys. Rev. Lett. **92**, 017401 (2004).

[25] S. Savasta and R. Girlanda, Phys. Rev. B **59**, 15409 (1999).

[26] V. Savona, L. C. Andreani, P. Schwendimann, and A. Quattropani, Solid State Commun. **93**, 733 (1995).

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

[28] C. Ciuti, P. Schwendimann, and A. Quattropani, Phys. Rev. B **63**, 041303(R) (2001); C. Ciuti, P. Schwendimann, and A. Quattropani, Semicond. Sci. Technol. **18**, S279 (2003).

[29] G. Rochat, C. Ciuti, V. Savona, C. Piermarocchi, A. Quattropani, and P. Schwendimann, Phys. Rev. B **61**, 13856 (2000).

[30] From the point of view of experiments by probing the quantum properties of the emitted radiation out of a SMC (our aim) in common structures (e.g., GaAs), the Rabi splitting amounts of a bunch of meV. As a consequence, in order to be safely off-resonance, the biexciton energy needs not to be more far in energy of some dozens of meV.

[31] R. Takayama, N. H. Kwong, I. Rumyantsev, M. Kuwata-Gonokami, and R. Binder, Eur. Phys. J. B **25**, 445 (2002).

[32] N. H. Kwong, R. Takayama, I. Rumyantsev, M. Kuwata-Gonokami, and R. Binder, Phys. Rev. B **64**, 045316 (2001).

[33] S. Schumacher, N. H. Kwong, and R. Binder, Phys. Rev. B **76**, 245324 (2007).

[34] M. Combescot, O. Betbeder-Matibet, and R. Combescot, Phys. Rev. B **75**, 174305 (2007).

[35] O. Di Stefano, S. Savasta, and R. Girlanda, Phys. Rev. A **60**, 1614 (1999).

[36] C. C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Photons and Atoms: Introduction to Quantum Electrodynamics* (Wyley-Interscience, New York, 1989).

[37] T. Takagahara, Phys. Rev. B **31**, 6552 (1985).