Hierarchy of density matrices in coherent semiconductor optics

K. Victor, V. M. Axt, and A. Stahl
Institut für Theoretische Physik B, Rheinisch Westfälische Technische Hochschule Aachen, Sommerfeldstraße, 52056 Aachen, Germany
(Received 5 December 1994; revised manuscript received 17 February 1995)

The hierarchy of many-body density matrices describing the nonlinear optical response of semiconductors is studied. The analysis is restricted to the coherent electronic dynamics generated by the relevant Hamiltonian. The strength of the optical excitation is used as a perturbation parameter, allowing a controlled truncation of the hierarchy and a drastic reduction of the number of independent multipoint functions describing the response to any given order. Thus the many-body effects contributing to the nth order susceptibility $\chi^{(n)}$ are described by $[n+1/2]$ independent multipoint functions. As a consequence of the assumed coherence it turns out that all densitylike variables can be expressed in terms of transitionlike quantities. The proposed systematic treatment of Coulomb correlations is compared with the conventional random-phase approximation factorization of many-body matrices.

I. INTRODUCTION

A considerable part of the work done in time-resolved nonlinear optics of semiconductors serves the purpose of studying the formation of resonant many-body structures such as excitons and the subsequent decay of these transient structures through relaxation and dephasing processes.[1–12] Although excitons are the most prominent structures, there is plenty of evidence that under suitable excitation conditions biexcitons and even higher electron-hole complexes do influence the observed signals.[5–13]

The pertinent theory describing the dynamics of the semiconductor electrons under the influence of the driving laser field has the structure of an open hierarchy of multipoint functions. As those, one may use either a set of single-time reduced density matrices[14] or the corresponding multitime Green functions.[15–17] We shall concentrate here on the density-matrix approach.

Whenever one is dealing with a hierarchy of dynamical objects one is facing the problem of finding an appropriate termination procedure. Since the hierarchy of electronic multipoint functions is generated by the Coulomb interaction, at first sight it seems plausible to seek a termination based on orders in the interaction. But due to the long-range character of the Coulomb potential this does not work, at least with the unscreened potential. A way out of this dilemma is the classification of the many-body effects as either screening or scattering, treating the screening nonperturbatively.[18–20]

In our approach we shall not use the interparticle interaction as a perturbation parameter. Inspired by the classification of the nonlinear optical response according to orders in the driving field, we base our truncation scheme on the smallness of the optical excitation. This is a well suited parameter for the classification of many-body effects because the unexcited crystal can be described as a zero-particle system using the electron-hole picture. All many-body correlations then are generated by the optical excitation.

The widely used random-phase approximation (RPA) factorization approximates the many-body correlations by the use of single-particle densities, i.e., two-point functions only. In this way one is lead to the semiconductor Bloch equations.[21–24] Despite its success in many cases, this approach nevertheless is questionable because it neglects higher correlations in an uncontrollable way, correlations that in many cases are important. In contrast, the controlled truncation scheme based on powers of the excitation strength as it was presented in a recent paper by two of us[25] opens a way to describe the many-body correlations by the use of a finite set of few-point density matrices with an error that is of fixed but arbitrary order in the excitation strength. The result is a closed set of equations, which is a natural generalization of the semiconductor Bloch equations. In the present paper we further extend this method and analyze the internal structure of the hierarchy. We explore exact interrelations between specific classes of density matrices, allowing us to define the minimal set of dynamical variables that are necessary for the description of $\chi^{(n)}$ experiments. When we denote our equations as exact up to any prescribed order this is to be understood in the restricted sense of the coherent dynamics generated by the pertinent Hamiltonian. The problem of a similarly systematic treatment of the coupling to a heat bath is not discussed in this paper.

Our paper is organized as follows. After having specified in Sec. II our semiconductor model, we discuss the structure of the resulting hierarchy of density matrices in Sec. III and give a geometrical visualization in the form of an open pyramid. The central theorems of the paper are presented in Sec. IV and discussed on an intuitive level. The proofs are deferred to the Appendixes. The time evolution of the system state is expanded according to orders in the optical field. The connection to the many-body character of higher-order correlation functions is worked out in detail. Theorems that allow us to reduce the number of independent density matrices in a $\chi^{(n)}$ calculation are derived. Two of these theorems are generalizations of contraction and factorization rules known to third order of the optical field.[25,26] The present

theorems hold to arbitrary order. Section V is devoted to a discussion.

II. MODEL HAMILTONIAN

Our model relies on two main assumptions. First, the initial state of our system has to be the unexcited ground state $|0\rangle$

$$\hat{d}_i|0\rangle=0, \quad \hat{c}_i|0\rangle=0 , \tag{2.1}$$

where $\hat{d}_i(\hat{c}_i)$ is the hole (electron) annihilator for Wannier states i. The index i summarizes band and site information. Thus summations over Wannier states i and j involve a three-dimensional integration over the crystal volume and a summation over all bands of either valence or conduction type depending on whether the corresponding operators represent electrons or holes. The assumption concerning the initial state implies that the optical excitation should vanish for $t\rightarrow-\infty$. The second assumption is that the material dynamics is generated by the Hamiltonian

$$H=H_0+H_{EP} , \tag{2.2}$$

with

$$H_0=H_{\text{band}}+H_\Phi+H_{\text{int}} ,$$

$$H_{\text{band}}=\sum_{i,j} T_{ij}^c\hat{c}_i^\dagger\hat{c}_j+\sum_{i,j} T_{ij}^v\hat{d}_i^\dagger\hat{d}_j ,$$

$$H_\Phi=e\left[\sum_i \hat{d}_i^\dagger\hat{d}_i\Phi_i^v-\sum_i \hat{c}_i^\dagger\hat{c}_i\Phi_i^c\right] ,$$

$$H_{\text{int}}=\frac{1}{2}\sum_{i,j}(\hat{c}_i^\dagger\hat{c}_i-\hat{d}_i^\dagger\hat{d}_i)V_{ij}(\hat{c}_i^\dagger\hat{c}_j-\hat{d}_j^\dagger\hat{d}_j) ,$$

and

$$H_{EP}=H_{EP}^{(+)}+\text{H.c.} ,$$

$$H_{EP}^{(+)}=\sum_{i,j} M_{ij}\hat{d}_i^\dagger\hat{c}_j^\dagger E_{(ij)}(t) .$$

H_{band} is the undisturbed single-particle energy of the system and H_Φ represents the interaction with an external potential (including the spatial dependence of the band edges in structured sample). Coulomb interactions of a monopole-monopole type[14,21,27] are taken into account by H_{int}, with the on-site potential $V_{ii}=0$. The real-valued optical field $E_{(ij)}(t)$ at a location between the sites i and j couples to the system via the electric dipole interaction H_{EP}. This Hamiltonian has been used successfully by many authors to describe optical processes in semiconductors.[17,19,20,23,24,28,29]

In the subsequent analysis mainly one feature of the above Hamiltonian will be used: The only part of H that does not conserve the total number of conduction electrons and the total number of holes separately is the coupling to the light field H_{EP}. More precisely, this means

$$[\hat{n}_{c/v},H_0]=0, \quad [\hat{n}_{c/v},H_{EP}^{(+)}]=H_{EP}^{(+)} \tag{2.3}$$

with

$$\hat{n}_c:=\sum_i \hat{c}_i^\dagger\hat{c}_i, \quad \hat{n}_v:=\sum_i \hat{d}_i^\dagger\hat{d}_i \tag{2.4}$$

III. HIERARCHY OF DENSITY MATRICES

The relevant observables in optical experiments are the interband polarization P and the charge density ρ. These observables are derived from the single-particle density matrices $Y_2^1=\langle\hat{d}_1\hat{c}_2\rangle$, $C_{12}=\langle\hat{c}_1^\dagger\hat{c}_2\rangle$, and $D^{12}=\langle\hat{d}_1^\dagger\hat{d}_2\rangle$:

$$P(r_1,t)=\int_{r_1-r_i} M_{i1}^* Y_i^1+\text{c.c.} , \tag{3.1}$$

$$\rho(r_1,t)=-e(C_{11}-D^{11}) . \tag{3.2}$$

Unfortunately, the equations of motion for Y, C, and D do not form a closed set. Instead they are coupled to an infinite hierarchy of higher-order density matrices. It will turn out to be useful to introduce a notation for arbitrary density matrices by defining the operators ($n>0$)

$$\begin{aligned}
&\hat{Y}[n]_{(i)_n}^{(j)_n}:=\hat{d}_{j_1}\hat{c}_{i_1}\cdots\hat{d}_{j_n}\hat{c}_{i_n} ,\\
&\hat{c}[n]_{(i)_n}:=\hat{c}_{i_1}\cdots\hat{c}_{i_n} ,\\
&\hat{Y}[-n]_{(i)_n}^{(j)_n}:=\hat{c}_{j_1}\hat{d}_{i_1}\cdots\hat{c}_{j_n}\hat{d}_{i_n} ,\\
&\hat{c}[-n]_{(i)_n}:=\hat{d}_{i_1}\cdots\hat{d}_{i_n} ,\\
&\hat{Y}[0]=\hat{c}[0]:=\hat{1} .
\end{aligned} \tag{3.3}$$

Therewith, an arbitrary normally ordered density matrix can be written in the form

$$\langle\hat{c}^\dagger[n_u]_\lambda\hat{Y}^\dagger[n_p^+]_\nu^{\nu'}\hat{Y}[n_p^-]_\mu^{\mu'}\hat{c}[n_u]_\kappa\rangle . \tag{3.4}$$

We have only considered operators conserving the total particle number in the electron picture. Obviously only these operators are coupled to Y, C, and D by the Hamiltonian. The operator in (3.4) is characterized by three integers: n_p^- pairs are annihilated at locations that are summarized in the pair of index sets (μ,μ'), a number n_p^+ of pairs is created at the sites (ν,ν'), and a number n_u of unpaired electrons (or holes for negative n_u) is transferred from the Wannier sites κ to λ. Thus the operator is of mixed transition and transport (density) type. The infinite set of density matrices can most conveniently be visualized if we arrange them in a space spanned by n_u and two other integers defined as

$$\begin{aligned}
&m:=n_p^++n_p^-+2|n_u|\in\{1,\ldots,\infty\} ,\\
&n_g:=n_p^--n_p^+\in\{-m,-m+2,\ldots,m\} ,\\
&n_u\in\left\{-\frac{m-|n_g|}{2},\ldots,\frac{m-|n_g|}{2}\right\} .
\end{aligned} \tag{3.5}$$

The *majority number* m in (3.5) is either the total number of electron operators or hole operators depending on which is larger. The *gap order* n_g is the number of effectively annihilated pairs. (3.4) has resonances near a frequency of n_g times the gap frequency. The *number of unpaired particles* n_u by its sign expresses the particle type, i.e., electron or hole.

The density matrices of type (3.4) form an open pyramid in the (m,n_g,n_u) space centered along the m axis (see Fig. 1). The points in this pyramid of density matrices are coupled to their next neighbors by the optical field E

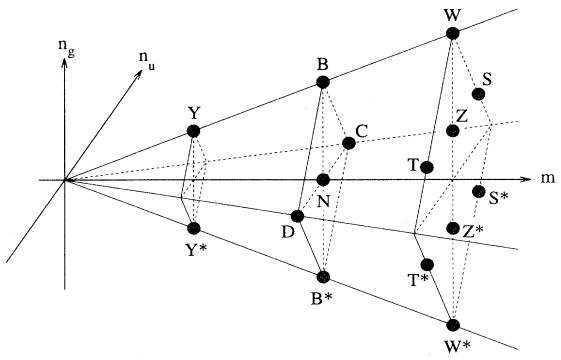

FIG. 1. Hierarchy of density matrices up to $m=3$. The symbols are defined as $Y:=\langle \hat{d}\hat{c}\rangle$, $C:=\langle \hat{c}^\dagger\hat{c}\rangle$, $D:=\langle \hat{d}^\dagger\hat{d}\rangle$, $N:=\langle \hat{c}^\dagger\hat{d}^\dagger\hat{d}\hat{c}\rangle$, $B:=\langle \hat{d}\hat{c}\hat{d}\hat{c}\rangle$, $S:=\langle \hat{c}^\dagger\hat{c}\hat{d}\hat{c}\rangle$, $T:=\langle \hat{d}^\dagger\hat{d}\hat{d}\hat{c}\rangle$, $W:=\langle \hat{d}\hat{c}\hat{d}\hat{c}\hat{d}\hat{c}\rangle$, and $Z:=\langle \hat{c}^\dagger\hat{d}^\dagger\hat{d}\hat{c}\hat{d}\hat{c}\rangle$. The lines are only guides to the eye.

and by the Coulomb interaction V. This means that the equation of motion for a density matrix $X(m,n_g,n_u)$ contains source terms of the form

$$
\begin{aligned}
&E\times X(m-1,n_g\pm1,n_u)\ ,\\
&E\times X(m-1,n_g\pm1,n_u+1)\ ,\\
&E\times X(m+1,n_g\pm1,n_u-1)\ ,\\
&V\times X(m+2,n_g,n_u-1)\ ,\\
&V\times X(m,n_g,n_u+1)
\end{aligned}
$$

for $n_u<0$,

$$
\begin{aligned}
&E\times X(m-1,n_g\pm1,0)\ ,\\
&E\times X(m+1,n_g\pm1,+1)\ ,\\
&E\times X(m+1,n_g\pm1,-1)\ ,\\
&V\times X(m+2,n_g\pm1)
\end{aligned}
\qquad (3.6)
$$

for $n_u=0$, and

$$
\begin{aligned}
&E\times X(m-1,n_g\pm1,n_u)\ ,\\
&E\times X(m-1,n_g\pm1,n_u-1)\ ,\\
&E\times X(m+1,n_g\pm1,n_u+1)\ ,\\
&V\times X(m+2,n_g,n_u+1)\ ,\\
&V\times X(m,n_g,n_u-1)
\end{aligned}
$$

for $n_u>0$. While the Coulomb interaction couples only variables within horizontal planes of the pyramid (with the same gap order), the optical field couples neighboring planes with $n_g-n_g'=\pm1$. Terms with the optical field E do not lead back from higher-order density matrices to the electrodynamically relevant quantities Y, C, and D, whereas the Coulomb interaction does. Thus an exact calculation of the electrodynamic observables requires us to deal with the whole infinite hierarchy of many-body density matrices.

IV. TRUNCATION, CONTRACTION, AND FACTORIZATION THEOREMS

In Sec. IV A we analyze the many-body character of the optically driven electronic system using the Schrödinger picture. This analysis is useful for the physical understanding and for the proof of various relations between density matrices. These relations, discussed in Sec. IV B, are the manifestation of the internal structure of the hierarchy pyramid and allow us to reduce the infinite set of many-body functions to a limited subset of functions relevant for $\chi^{(n)}$ calculations.

A. Connection between the order of perturbation and the many-body character of the system

We say that a system of many particles has a *many-body character,* if it is not possible to describe it using only one-particle states as is done in a Hartree-Fock treatment. The many-body character of the system is responsible for the hierarchy problem. While the initial state effectively is of zero-particle character in the electron-hole picture, the many-body character is generated through the action of the external optical field. According to H_{EP} the effect of applying the optical field once is either the creation or the annihilation of one electron-hole pair. This leads to a time evolution of the system state such that an expansion with respect to the optical field and a decomposition in Fock space into states with definite numbers of electrons and holes are not independent of each other.[11]

In order to make this connection explicit we expand the Schrödinger state of the system at time t according to time-dependent perturbation theory[30] in powers n of the external field E:

$$|t\rangle=\sum_n |t\rangle^{(n)} \quad \text{with } |t\rangle^{(n)}\propto E^n\ , \qquad (4.1)$$

$$|t\rangle^{(n)}=\frac{1}{i\hbar}\int_{-\infty}^{t} dt' e^{-(i/\hbar)H_0(t-t')}H_{EP}(t')|t'\rangle^{(n-1)}\ , \qquad (4.2)$$

$$|t\rangle^{(0)}=e^{-(i/\hbar)H_0 t}|0\rangle\ . \qquad (4.3)$$

A further decomposition of the state $|t\rangle^{(n)}$ into states $|n_c,n_v,n,t\rangle$ with definite numbers n_c and n_v of electrons and holes, respectively, is always possible:

$$|t\rangle^{(n)}=\sum_{n_c,n_v=0}^{\infty}|n_c,n_v,n,t\rangle \qquad (4.4)$$

with

$$\hat{n}_{c/v}|n_c,n_v,n,t\rangle=n_{c/v}|n_c,n_v,n,t\rangle\ . \qquad (4.5)$$

Using the two properties (2.3) and (2.1) of our Hamiltonian and the above recursion (4.2) and (4.3) we prove by induction (see Appendix A) the following theorem.

Theorem 1 (expansion theorem). At any time the Schrödinger state of the system can be decomposed into contributions of fixed order n in the optical field. Each of these contributions can be expressed in a basis of states with definite total numbers of electrons n_c and holes n_v, where the number of electrons always equals that of the

holes and is smaller than or equal to the order n. The difference between $n_p := n_c = n_v$ and n is even. Thus we can write

$$|t\rangle^{(n)} = \sum_{n_p = n, n-2, \ldots \geq 0} |n_p, n_p, n, t\rangle \ .$$

An equivalent formulation of the first part of the above theorem ($n_c = n_v \leq n$) has already been proved in Ref. 25. The extension ($n - n_p$ even) is easily understood on a qualitative basis. Any n_p-pair state can be thought of as being generated by the following sequence of processes. First the optical field is applied n_p times to create n_p pairs followed by $k \geq 0$ further creation processes and k annihilations. Thus any n_p-pair state is generated in a process of order $n_p + 2k$ in the driving field. Therefore a component with n_p pairs can only result in a contribution of order n if $n - n_p$ is even.

The principal physical meaning of the expansion theorem is that all charge carriers exist in electron-hole pairs and that the many-body character of the system can be classified according to an expansion in terms of the optical field. To nth order in the optical field "many-body character" means the presence of states with up to n pairs.

B. Consequences for the truncation and interrelations between density matrices

Expectation values of normally ordered n-particle operators identically vanish if the system is in a state with less than n particles (this is what normal order means). Because the number of particles in the states that are required to describe the system can be related to the order of perturbation, as has been shown in Sec. IV A, the expectation values of operators involving a large number of particle creators and annihilators will have vanishing values except for high orders in the optical field. Thus if one is interested in a fixed order of perturbation, e.g., a $\chi^{(3)}$ experiment, the hierarchy of density matrices can be truncated exactly at a distinct few-particle level, neglecting all many-body density matrices that are of higher order.

Theorem 2 (truncation theorem). A given density matrix $X(m, n_g, n_u)$ as defined by (3.5) is at least of order m in the optical field. Furthermore X is either even or odd in the electrical field:

$$X = \sum_{l=0}^{\infty} X^{(m+2l)} \quad \text{with } X^{(k)} \propto E^k \ .$$

This theorem has been used in Ref. 25 to give a rigorous truncation of the hierarchy at the $\chi^{(3)}$ level. The classification of density matrices into those of even and those of odd order in the optical field is new. A similar truncation scheme has been applied to a system of Frenkel excitons.[31] The proof of the truncation theorem (given in Appendix B) consists of a straightforward application of the expansion theorem.

The truncation theorem limits the set of $\chi^{(n)}$-relevant variables to those density matrices that contain up to n electron and n hole operators. In the visualization of Fig. 1 this means that the open pyramid is terminated at $m = n$. Up to order n only those multipoint functions that are within the pyramid $m \leq n$ will be excited. But not all these functions will affect the $\chi^{(n)}$-type electromagnetic response of the single-particle functions. For example, the triexcitonic amplitude $W := \langle \hat{c}\hat{c}\hat{d}\hat{c}\hat{d}\hat{c} \rangle$ is of third order in the optical field, but W does not couple back to Y, C, or D within $\chi^{(3)}$ via the sources (3.6) (see Ref. 25). Although higher-order density matrices $X(m, n_g, n_u)$ are excited within the order m in the optical field, they do not influence the electrodynamics except through the source terms (3.6) of the equations of motion of Y, C, and D. For most of them this requires convolutions with other functions, leading to additional powers in the optical field.

This situation is illustrated in Fig. 2 showing the projection of the density-matrix pyramid onto the plane spanned by the majority number m and the gap order n_g. Figure 2(a) displays the coupling of density matrices involving the optical field E. This coupling is from left to right or bidirectional, e.g., $E \times Y$ is a source in the equations of motion for the biexcitonic transition B and for the electronic density C and $E \times C$ in turn is a blocking type source for the exciton transition Y (phase-space filling). In contrast, the Coulomb coupling displayed in Fig. 2(b) is unidirectional from right to left; only density

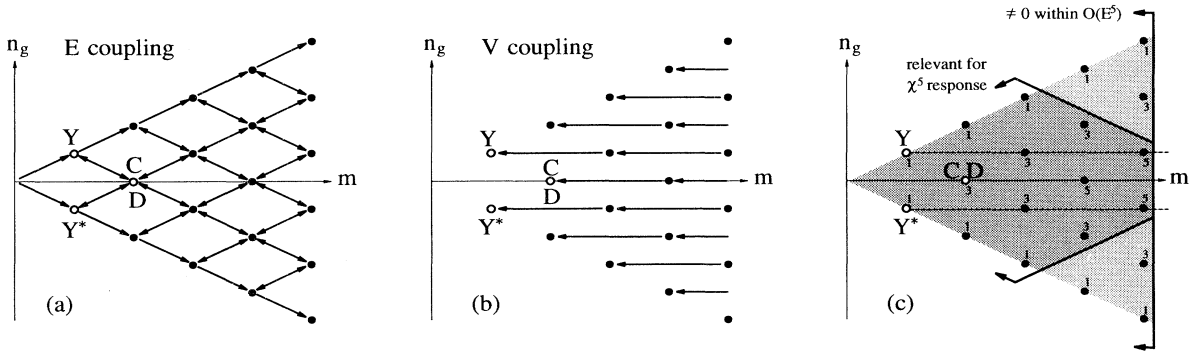

FIG. 2. (a) In the projection of the density matrix pyramid onto the plane spanned by m and n_g, the couplings between different variables induced by the optical field form a bidirectional network. (b) The Coulomb potential couples only variables with the same gap order n_g. It never leads to source terms with a lower majority number m than the driven density matrix. As stated in Theorem 3, this restricts the set of $\chi^{(n)}$-relevant variables, as displayed in (c) for the case $n = 5$. The number of density matrices with different numbers of unpaired particles n_u, which in this projection are represented by one point, is indicated.

matrices with higher (or identical) m are Coulomb sources to other variables. For $t \to -\infty$ all variables are zero. As only Y has a source linear in E not involving any other density matrix, the variables in the hierarchy are excited successively from left to right due to the action of the optical field. The iterative solution of the equations of motion resembles the movement of a draughtsman on a gameboard, where each step of the draughtsman corresponds to one iteration. Using the network of E sources in Fig. 2(a), a draughtsman starting at the left corner needs n steps to reach the vertical plane with $m=n$, each step at the expense of one order of the optical field. This actually is the truncation theorem. At the point (m,n_g,n_u) that draughtsman acquires specific information about the many-body correlations described by the associated variable $X(m,n_g,n_u)$. In order to inform the experimentalist, who can only detect electromagnetic signals, the draughtsman has to carry his knowledge back to one of the points Y, Y^*, C, or D. To do this, the player may again use the streets of Fig. 2(a), but each step would then introduce an additional order in the optical field. On the return journey, however, the player can alternatively use the "free-ticket" lines of the Coulomb sources shown in Fig. 2(b). These lines do not increase the order of the optical field, but it should be noted that they connect only horizontal planes with constant n_g. Thus at least $||n_g|-1|$ steps must be taken on the E network to reach the planes of interband polarizations (Y,Y^*). Likewise $|n_g|$ steps are required to influence the charge densities (C,D). These considerations are summarized in the following theorem.

Theorem 3 (electrodynamic response theorem). A $\chi^{(n)}$ calculation of the electromagnetic response to an optical field only requires the evaluation of the density matrices $X(m,n_g,n_u)$ with $m+\max(|n_g|-1,0)\le n$ to orders $n-\max(|n_g|-1,0)$ in the optical field.

Figure 2(c) shows the region of density matrices with $m+\max(|n_g|-1,0)\le 5$ relevant for the $\chi^{(5)}$ interband-polarization response of the semiconductor. Thus 22 dynamical variables remain for the description of $\chi^{(5)}$ experiments and eight remain for the case of $\chi^{(3)}$ (see Ref. 25).

Still it would be practically impossible to solve all these equations. But, fortunately, the relevant variables are interrelated by a number of identities. In Ref. 26 it was shown that for the $\chi^{(3)}$ case the set of independent variables can be reduced to the excitonic transition Y and the biexcitonic transition $B:=\langle \hat{d}\hat{c}\hat{d}\hat{c}\rangle$. The identities needed to express the other six variables by Y and B were found by inspection of the equations of motion in Ref. 26. Here we generalize these identities by the subsequent two theorems.

While the truncation theorem expresses the fact that for each generation (annihilation) of carriers the field has to be applied once, the theorem does not yet make use of the feature that in each process a pair is involved. In a system, however, where all carriers are generated (annihilated) in pairs of electrons and holes the process of moving an electron from site x to x' as described by the variable $C_{x'x}:=\langle \hat{c}_x^\dagger \hat{c}_x\rangle$ is equivalent to a process within an exciton, where in addition a hole is present at site i (see Fig. 3). Of course one has to sum over all possible hole sites i. Higher orders in the optical field call for corrections due to processes within biexcitons, triexcitons, etc. This is the essence of the following theorem proved in Appendix C.

Theorem 4 (contraction theorem). Density matrices with a nonvanishing number of unpaired particles $(n_u\neq 0)$ can be expressed by contractions of density matrices with the same gap order n_g and the same or higher majority number m containing only pair operators $(n_u=0)$:

$$\langle \hat{c}^\dagger[n_u]_\lambda \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{Y}[n_p^-]_\mu^{\mu'} \hat{c}[n_u]_\kappa\rangle = \sum_{k=0}^{l} g_{|n_u|,k} \sum_{(i)_{|n_u|},(j)_k,(j')_k} \langle \hat{Y}^\dagger[n_u]_\lambda^{(i)_{|n_u|}} \hat{Y}^\dagger[k+n_p^+]_{(j)_k\cdot\nu}^{(j')_k\cdot\nu'} \hat{Y}[k+n_p^-]_{(j')_k\cdot\mu}^{(j')_k\cdot\mu'} \hat{Y}[n_u]_\kappa^{(i)_{|n_u|}}\rangle \,.$$

$$+O(E^{m+2+2l}) \quad \text{with } m=n_p^+ + n_p^- + 2|n_u| \,.$$

The rule for a successive calculation of the coefficients $g_{n,k}$ reads

$$g_{n,k}=\frac{1}{(k+n)!k!}-\sum_{k'=0}^{k-1}\frac{g_{n,k'}}{(k-k')!^2}, \quad g_{n,0}=\frac{1}{n!} \,.$$

In the formulation of the theorem the dot operation means concatenation of two multi-indices to one. The contraction theorem reduces the three-dimensional hierarchy pyramid in Fig. 1 to the central vertical sheet with $n_u=0$. This is a consequence of the expansion theorem which states that in the three-dimensional decomposition of the Schrödinger state only the contributions with equal electron and hole numbers do not vanish. Similar to the expansion of the Schrödinger state with respect to n and n_p, the remaining density matrices form a two-dimensional set. The third dimension is eliminated because of the pairwise generation of carriers.

While the contraction theorem connects variables with comparable free oscillation frequencies (with the same gap order n_g), in Ref. 25 a second type of identity was also derived, establishing a relation between slow and fast

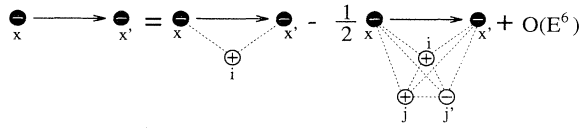

FIG. 3. Contraction theorem ($n_u=1$, $l=1$, $n_p^+=n_p^-=0$): As electrons always belong to electron-hole pairs, the transport of an electron can be thought to be composed of contributions belonging to an internal electron transport in an exciton and in a biexciton to $O(E^4)$.

variables. In particular it was shown that the exciton density N can be decomposed to lowest order into a product of transition amplitudes

$$N_{14}^{23}:=\langle \hat{c}_1^\dagger \hat{d}_2^\dagger \hat{d}_3 \hat{c}_4 \rangle = Y_1^{*2} Y_4^3 + O(E^3) . \tag{4.6}$$

This factorization rule expresses the fact that excitons behave like bosons to lowest order in the particle number (and thus to lowest order in the optical field). If bosons, e.g., photons, are generated by a classical force as done with the excitons by the coupling to the optical field E, a coherent state is the result. This means that intensities can be expressed as the product of amplitudes. As long as the analogy to bosons holds, exciton densities can be expressed as the product of exciton transition amplitudes. A generalization of (4.6) to arbitrary order of the optical field has to consider deviations from the bosonic behavior that arise from phase-space filling and the Coulomb interactions. This is achieved by the following theorem.

Theorem 5 (factorization theorem). Let $\hat{P}_l$ be an operator acting in the subspace of states with equal numbers n of electrons and holes and at maximum l pairs ($n \leq 1$) as a projector to the ground state $|0\rangle$. All other states with definite particle number are assumed to be eigenstates of $\hat{P}_l$. Then the following factorization of density matrices with vanishing number of unpaired particles ($n_u=0$) holds:

$$\begin{aligned}
&\langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{Y}[n_p^-]_\mu^{\mu'} \rangle \\
&\quad = \langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \rangle \langle \hat{Y}[n_p^-]_\mu^{\mu'} \rangle \\
&\qquad + \sum_{k=1}^{l} p_k \sum_{(i)_k,(j)_k} \left(\langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \rangle \langle \hat{Y}^\dagger[k]_{(i)_k}^{(j)_k} \hat{Y}[k+n_p^-]_{(i)_k \cdot \mu}^{(j)_k \cdot \mu'} \rangle - \langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{Y}[n_p^-]_\mu^{\mu'} \rangle \langle \hat{Y}^\dagger[k]_{(i)_k}^{(j)_k} \hat{Y}[k]_{(i)_k}^{(j)_k} \rangle \right) \\
&\qquad + \sum_{k=1}^{l} p_k \sum_{(i)_k,(j)_k} \sum_{k'=0}^{l-k} p_{k'} \sum_{(i')_{k'},(j')_{k'}} \left(\langle \hat{Y}^\dagger[k+n_p^+]_{(i)_k \cdot \nu}^{(j)_k \cdot \nu'} \hat{Y}[k]_{(i)_k}^{(j)_k} \rangle \langle \hat{Y}^\dagger[k']_{(i')_{k'}}^{(j')_{k'}} \hat{Y}[k'+n_p^-]_{(i')_{k'} \cdot \mu}^{(j')_{k'} \cdot \mu'} \rangle \right. \\
&\qquad\qquad \left. - \langle \hat{Y}^\dagger[k+n_p^+]_{(i)_k \cdot \nu}^{(j)_k \cdot \nu'} \hat{Y}[k+n_p^-]_{(i)_k \cdot \mu}^{(j)_k \cdot \mu'} \rangle \langle \hat{Y}^\dagger[k']_{(i')_{k'}}^{(j')_{k'}} \hat{Y}[k']_{(i')_{k'}}^{(j')_{k'}} \rangle \right) \\
&\qquad\qquad + O(E^{n_p^+ + n_p^- + 2 + 2l}) .
\end{aligned} \tag{4.10}$$

For $l=0$ we get the factorization rules to lowest order

$$\langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{Y}[n_p^-]_\mu^{\mu'} \rangle = \langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \rangle \langle \hat{Y}[n_p^-]_\mu^{\mu'} \rangle + O(E^{n_p^+ + n_p^- + 2}) . \tag{4.11}$$

The rule (4.6) is of this type. For higher accuracies l, the sums in (4.10) involve partially densitylike terms, i.e., expectation values that contain annihilators as well as creators. These can be eliminated applying (4.10) iteratively with a lower accuracy, For $l=1$ one obtains

$$\begin{aligned}
&\langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{Y}[n_p^-]_\mu^{\mu'} \rangle \\
&\quad = \langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \rangle \langle \hat{Y}[n_p^-]_\mu^{\mu'} \rangle \\
&\qquad + \sum_{i,j} \langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} * \hat{Y}^\dagger[1]_i^j \rangle \langle \hat{Y}[1]_i^j * \hat{Y}[n_p^-]_\mu^{\mu'} \rangle \\
&\qquad + O(E^{n_p^+ + n_p^- + 4}) ,
\end{aligned} \tag{4.12}$$

$$\begin{aligned}
&\langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{P}_l \hat{Y}[n_p^-]_\mu^{\mu'} \rangle \langle \hat{P}_l \rangle \\
&\quad = \langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{P}_l \rangle \langle \hat{P}_l \hat{Y}[n_p^-]_\mu^{\mu'} \rangle + O(E^{n_p^+ + n_p^- + 2 + 2l}) .
\end{aligned} \tag{4.7}$$

A convenient choice for the projector $\hat{P}_l$ is

$$\hat{P}_l := \sum_{k=0}^{l} p_k \sum_{(i)_k,(j)_k} \hat{Y}^\dagger[k]_{(i)_k}^{(j)_k} \hat{Y}[k]_{(i)_k}^{(j)_k} , \tag{4.8}$$

where the coefficients p_k can be calculated recursively from

$$p_k = -\sum_{k'=0}^{k-1} \frac{p_{k'}}{(k-k')!^2} , \quad p_0 = 1 .$$

The proof of this factorization and of the required properties of $\hat{P}_l$ as constructed above is given in Appendix D.

The projectors $\hat{P}_l$ compensate deviations from the boson character of multiexciton systems. Obviously, the integer l that determines the extension of the subspace where $\hat{P}_l$ acts as a projector has the role of an index of accuracy in (4.7). The first two projectors are given by

$$\hat{P}_0 := = \hat{1}, \quad \hat{P}_1 := \hat{1} - \sum_{i,j} \hat{Y}^\dagger[1]_i^j \hat{Y}[1]_i^j . \tag{4.9}$$

The theorem will most often be used in a rearranged form, where the representation (4.8) of $\hat{P}_l$ has been inserted:

where we have used the notation

$$\langle \hat{A} * \hat{B} \rangle := \langle \hat{A}\hat{B} \rangle - \langle \hat{A} \rangle \langle \hat{B} \rangle .$$

Equation (4.12) is illustrated in Fig. 4. For arbitrary l we express all partially densitylike variables $\langle \hat{Y}^\dagger[n_p^+] \hat{Y}[n_p^-] \rangle$ by a sum of products of transition amplitudes $\langle \hat{Y}^\dagger[k_q^+] \rangle$ and $\langle \hat{Y}[k_q^-] \rangle$ with $k_q^+ \leq n_p^+ + l$ and $k_q^- \leq n_p^- + l$. Using (4.10) iteratively we can determine a coefficient a_q for each term (labeled by q). The expectation value of the operator product then takes on the following form:

$$\langle Y^\dagger[n_p^+] Y[n_p^-] \rangle = \sum_q a_q \prod_{k_q^+} \langle Y^\dagger[k_q^+] \rangle \prod_{k_q^-} \langle Y[k_q^-] \rangle + O(E^{n_p^+ + n_p^- + 2l + 2}) \tag{4.13}$$

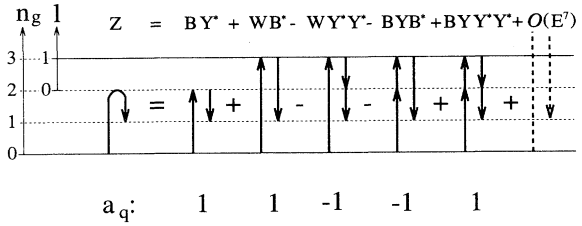

FIG. 4. For $l=1$ Eq. (4.13) reduces to (4.12). The figure illustrates the factorization of $Z=\langle \hat{Y}^*[1]\hat{Y}[2]\rangle$ as an example for (4.12). Each unit of an upward (downward) arrow represents one exciton annihilator (creator) and increments the order of the optical field by one.

with

$$k_q^+ \le n_p^+ + l, \quad k_q^- \le n_p^- + l \quad \text{for all } k_q^+, k_q^- \ .$$

The sum of all gap orders in each product is equal to the gap order $n_p^- - n_p^+$ of the unfactorized variable

$$\sum k_q^- - \sum k_q^+ = n_p^- - n_p^+ \ . \tag{4.14}$$

The proof of (4.13) and (4.14) is straightforward, using (4.10) and complete induction in l.

Thus to a given order in the optical field, the whole pyramid of density matrices as illustrated in Fig. 1 can be expressed by the upper edge with $n_u=0$ (contraction theorem) and $n_g=m$ (factorization theorem). As only the first n of these transition amplitudes have nonvanishing values within $O(E^n)$ (truncation theorem), only these have a chance to contribute to a $\chi^{(n)}$ signal. According to Theorem 3 (electrodynamic response theorem) not all of them actually do. In Appendix E it is shown that the use of the contraction and factorization theorems does not conflict with Theorem 3, i.e., that the application of these theorems to a variable $X(m,n_g,n_u)$ with $m+\max(|n_g|-1,0)\le n$ with accuracy $O(E^{n-\max(|n_g|-1,0)})$ does not involve density matrices $X'(m',n_g',n_u')$ with $m'+\max(|n_g'|-1,0)>n$ or accuracies higher than $O(E^{n-\max(|n_g|-1,0)})$. This leads to the following concluding theorem.

Theorem 6 (minimal set of $\chi^{(n)}$ relevant variables). Using the contraction theorem a $\chi^{(n)}$ calculation of the electromagnetic material polarizations requires only the evaluation of the density matrices $X(m,n_g,0)$ with $m+\max(|n_g|-1,0)\le n$ to orders $n-\max(|n_g|-1,0)$ in the optical field. With the help of the contraction and factorization theorems it is possible to restrict the set of independent variables for which the equations of motion have to be solved to the transition densities $X(m,m,0)=Y[m]$ with $2m-1\le n$, which have to be determined up to order $n-m+1$.

V. DISCUSSION

In previous works the theory has been worked out in detail in a $\chi^{(3)}$ context. These applications are briefly reviewed in Sec. V A. As a first extension we present the dynamics relevant for the coherent $\chi^{(5)}$ response. In addition, we analyze the relation between the present theorems that provide an exact treatment of the Coulomb interaction and the usual RPA factorization of many-body density matrices. The paper closes with an outlook where the influence of dephasing processes on the validity of the theorems is discussed.

A. Applications to $\chi^{(3)}$

The method has been worked out in some detail up to order E^3 and applied successfully to two typical experiments.[26,32]

(i) The dynamical Stark effect in CuCl exhibits an anomalous behavior attributed to the resonance with the biexcitonic bound state.[7] It is clear that for a description of such an effect in terms of density matrices one must go beyond the standard RPA factorization on the two-point level. A treatment along the lines described in the present paper based on the functions Y and B has been shown to nicely reproduce all experimental features.[26]

(ii) The second example where our method has been successful in explaining an experimental result not accessible to an RPA-type calculation is the polarization dependence of a four-wave-mixing signal that has its origin in the biexcitonic continuum.[32]

B. Outline of the $\chi^{(5)}$-relevant dynamics

According to the concluding theorem of Sec. IV B, a $\chi^{(5)}$ calculation requires the solution of the equations of motion for the excitonic transition $Y=Y[1]$ in $O(E^5)$, for the biexcitonic transition $B=Y[2]$ in $O(E^4)$, and for the triexcitonic transition $W=Y[3]$ in $O(E^3)$. From the Hamiltonian (2.2) it is straightforward to derive the equations of motion for $Y[m]$:

$$\{-i\hbar\partial_t+\hbar\Omega[m]\}Y[m]^{(j)_m}_{(i)_m}=S_E[m]^{(j)_m}_{(i)_m}+S_C[m]^{(j)_m}_{(i)_m} \tag{5.1}$$

with

$$\hbar\Omega[m]Y[m]^{(j)_m}_{(i)_m}=\sum_{k=1}^{m}\left\{\sum_{i'}T^c_{i_k i'}Y[m]^{(j)_m}_{i_1\cdots i_{k-1}i'i_{k+1}\cdots i_m}+\sum_{j'}T^v_{j_k j'}Y[m]^{j_1\cdots j_{k-1}j'j_{k+1}\cdots j_m}_{(i)_m}\right.$$

$$\left.+e(\Phi^v_{j_k}-\Phi^c_{i_k})Y[m]^{(j)_m}_{(i)_m}+\sum_{l=1}^{m}\frac{1}{2}(V_{i_k i_l}+V_{j_k j_l}-2V_{j_k i_l})Y[m]^{(j)_m}_{(i)_m}\right\},$$

$$S_E[m]^{(j)_m}_{(i)_m}=\sum_{k=1}^{m}\left\{\sum_{l=1}^{m}(-1)^{k-l}M_{j_l i_k}E_{(j_l i_k)}Y[m-1]^{j_1\cdots j_{l-1}j_{l+1}\cdots j_m}_{i_1\cdots i_{k-1}i_{k+1}\cdots i_m}-\sum_{i'}M_{j_k i'}E_{(j_k i')}\langle\hat{c}^\dagger_{i'}\hat{Y}[m-1]^{j_1\cdots j_{k-1}j_{k+1}\cdots j_m}_{i_1\cdots i_{k-1}i_{k+1}\cdots i_m}\hat{c}_{i_k}\rangle\right.$$
$$\left.-\sum_{j'}M_{j' i_k}E_{(j' i_k)}\langle\hat{d}^\dagger_{j'}\hat{Y}[m-1]^{j_1\cdots j_{k-1}j_{k+1}\cdots j_m}_{i_1\cdots i_{k-1}i_{k+1}\cdots i_m}\hat{d}_{j_k}\rangle\right\},$$

$$S_C[m]^{(j)_m}_{(i)_m}=\sum_{k=1}^{m}\left\{\sum_{j'}(V_{j' i_k}-V_{j' j_k})\langle\hat{d}^\dagger_{j'}\hat{Y}[m]^{(j)_m}_{(i)_m}\hat{d}_{j'}\rangle-\sum_{i'}(V_{i' i_k}-V_{i' j_k})\langle\hat{c}^\dagger_{i'}\hat{Y}[m]^{(j)_m}_{(i)_m}\hat{c}_{i'}\rangle\right\}.$$

We assume a symmetric interaction potential $V_{ij}=V_{ji}$. The sources of (5.1) involve the variables $\langle\hat{c}^\dagger\hat{Y}[m]\hat{c}\rangle$ and $\langle\hat{d}^\dagger\hat{Y}[m]\hat{d}\rangle$. In order to get a closed set of equations these variables have to be expressed by Y, B, and W using the contraction and factorization rules of Sec. IV B. Applying these rules to penultimate lowest order of the optical field yields

$$\langle\hat{c}^\dagger_1\hat{Y}[m]^{(j)_m}_{(i)_m}\hat{c}_2\rangle=\sum_{j'}Y^*[1]^{j'}_1Y[m+1]^{(j)_m\cdot j'}_{(i)_m\cdot 2}+\sum_{j',i'',j''}\{(\tfrac{1}{2}Y^*[2]^{j''j'}_{i''1}-Y^*[1]^{j''}_{i''}Y^*[1]^{j'}_1)Y[m+2]^{(j)_m\cdot j''j'}_{(i)_m\cdot i''2}$$
$$-(Y^*[2]^{j''j'}_{i''1}-Y^*[1]^{j''}_{i''}Y^*[1]^{j'}_1)Y[m+1]^{(j)_m\cdot j'}_{(i)_m\cdot 2}Y[1]^{j''}_{i''}\}+O(E^{m+6}). \tag{5.2}$$

A similar result can be obtained for $\langle\hat{d}^\dagger\hat{Y}[m]\hat{d}\rangle$. With $m=0$, Eq. (5.2) also gives the electron densities expressed by Y and B. This is necessary for the calculation of intraband polarizations within $\chi^{(4)}$.

The source terms for Y, B, and W become

$$S_E[Y]^1_2=M_{12}E_{(12)}-\sum_{i,j}Y^{*j}_i(M_{1i}E_{(1i)}Y^j_2+M_{j2}E_{(j2)}Y^1_i)-\sum_{i,j,i',j'}\{(\tfrac{1}{2}B^{*j'j}_{i'i}-Y^{*j'}_{i'}Y^{*j}_i)(M_{1i}E_{(1i)}B^{i'j}_{i'2}+M_{j2}E_{(j2)}B^{j'1}_{i'i})$$
$$-(B^{*j'j}_{i'i}-Y^{*j'}_{i'}Y^{*j}_i)(M_{1i}E_{(1i)}Y^{j'}_iY^j_2+M_{j2}E_{(j2)}Y^{j'}_iY^1_i)\}$$
$$+O(E^7), \tag{5.3}$$

$$S_C[Y]^1_2=\sum_{i,j}(V_{i1}-V_{i2}-V_{j1}+V_{j2})\left\{Y^{*j}_iB^{1j}_{2i}+\sum_{i',j'}[(\tfrac{1}{2}B^{*jj'}_{ii'}-Y^{*j}_iY^{*j'}_{i'})W^{1jj'}_{2ii'}-(B^{*jj'}_{ii'}-Y^{*j}_iY^{*j'}_{i'})Y^{j'}_iB^{1j}_{2i}]\right\}+O(E^7), \tag{5.4}$$

$$S_E[B]^{13}_{24}=M_{12}E_{(12)}Y^3_4-M_{14}E_{(14)}Y^3_2-M_{32}E_{(32)}Y^1_4+M_{34}E_{(34)}Y^1_2$$
$$-\sum_{i,j}Y^{*j}_i(M_{1i}E_{(1i)}B^{j3}_{24}+M_{3i}E_{(3i)}B^{1j}_{24}+M_{2j}E_{(2j)}B^{13}_{i4}+M_{4j}E_{(4j)}B^{13}_{2i})+O(E^6), \tag{5.5}$$

$$S_C[B]^{13}_{24}=\sum_{i,j}Y^{*j}_iW^{13j}_{24i}(V_{i1}-V_{i2}+V_{i3}-V_{i4}-V_{j1}+V_{j2}-V_{j3}+V_{j4})+O(E^6), \tag{5.6}$$

$$S_E[W]^{135}_{246}=M_{12}E_{(12)}B^{35}_{46}-M_{14}E_{(14)}B^{35}_{26}+M_{16}E_{(16)}B^{35}_{24}-M_{32}E_{(32)}B^{15}_{46}+M_{34}E_{(34)}B^{15}_{26}$$
$$-M_{36}E_{(36)}B^{15}_{24}+M_{52}E_{(52)}B^{13}_{46}-M_{54}E_{(54)}B^{13}_{26}+M_{56}E_{(56)}B^{13}_{24}+O(E^5), \tag{5.7}$$

$$S_C[W]^{135}_{246}=O(E^5). \tag{5.8}$$

Equation (5.1) for $m=1,2,3$, together with (5.3)–(5.8), forms a closed set of three coupled equations for Y, B, and W that should be solved for the exact description of coherent $\chi^{(5)}$ experiments. Calculations of that kind[33] with the goal to explain exciton-biexciton beats observed in both the decay and the rising regime of time integrated four-wave mixing or six-wave mixing show promising results.

C. Comparison with the RPA factorization

In order to illustrate the physical meaning of the above theorems we compare our truncation of the hierarchy with the standard RPA approach. The lowest-order RPA treatment amounts of the truncation of the hierarchy by a decomposition of four-point functions in sums of products of the two-point functions Y, C, and D. From the k-space version of the RPA theory it is well known that the dynamics in homogeneous, fully coherent systems is constrained by the conservation of the Bloch sphere for each k. The conservation law holds for each k individually, although in a RPA treatment different k states are coupled by the Coulomb interaction. In the case of a two-band semiconductor the conservation law reads[34,35]

$$(1-2C_k)^2+4|Y_k|^2=1. \tag{5.9}$$

This rule is commonly used to reduce the number of independent variables in the description of coherent phe-

nomena.[34] In particular it opens a way to eliminate the slow variable C in favor of the fast variable Y. In our more general truncation scheme an analogous reduction of variables is achieved by combining contraction and factorization rules. They thus provide a natural generalization of the Bloch sphere conservation. This generalization introduces no approximations with respect to the Coulomb interaction and is not restricted to translationally invariant systems. Therefore it can be applied to the important case of semiconductor heterostructures and interfaces.

It has to be expected that our representation of C reduces to the RPA result when the many-body correlations contained in the higher-order density matrices $Y[n]$ are neglected. Indeed, using (5.2), keeping all terms up to $O(E^4)$ and replacing the biexcitonic transition density B by its RPA counterpart we find

$$C_{12}=\sum_j Y^{*j}_1 Y^j_2+\sum_{i,j,j'} Y^{*j}_i Y^{*j'}_1 Y^j_2 Y^{j'}_i+O(E^6)\ . \tag{5.10}$$

Noting that in homogeneous systems the two-point functions will depend only on the difference of their real-space coordinates, the summations become convolutions that factorize under Fourier transformation. Thus in k space we obtain

$$C_k=|Y_k|^2+|Y_k|^4+O(E^6)\ . \tag{5.11}$$

This is equivalent to (5.9) when terms higher than $O(E^4)$ are neglected.

Besides the electronic density C, all four-point functions can be expressed up to fourth order by the excitonic and the biexcitonic transition densities Y and B. By applying the RPA decoupling scheme to B in these relations we recover the corresponding RPA factorization for the respective four-point function. To illustrate this statement we consider the excitonic occupation density $N:=\langle \hat{c}^\dagger \hat{d}^\dagger \hat{d}\hat{c}\rangle$. Using the contraction and factorization theorems we get

$$\begin{aligned} N^{23}_{14} &\xrightarrow{\text{RPA for } B} Y^{*2}_1 Y^3_4+\sum_{j,l} Y^{*2}_j Y^{*l}_1 Y^l_4 Y^3_j+O(E^6) \\ &= Y^{*2}_1 Y^3_4+C_{14}D^{23}+O(E^6)\ . \end{aligned} \tag{5.12}$$

Thus in the low excitation regime, the neglect of four-particle correlations contained in the biexcitonic transition density B necessarily implies the RPA decomposition of all other four-point functions. Therefore we conclude that in the low excitation limit all deviations from the RPA behavior in our system are described by the difference between B and its RPA decomposition. This difference includes biexcitonic bound states as well as exciton-exciton interactions included in the corresponding scattering continuum.

D. Conclusion and outlook

In the present paper we have concentrated on the internal structure of the hierarchy of density matrices as it is dynamically induced in coherent nonlinear optics of semiconductors and we have proved a number of generally valid theorems. How the method works in practice has been demonstrated elsewhere.[26,32,33] We believe that at least up to order E^5 the systematic treatment of the hierarchy of density matrices can be worked out to become a reliable tool for the analysis of optical experiments with semiconductors. We expect from our experience that in the development of such a theory the proper treatment of continuum states will play a decisive role. Thus, e.g., the $\chi^{(3)}$ theory of four-wave mixing[32] (FWM) exhibits a strong influence of the exciton-exciton scattering continuum.

A discussion on which the results obtained in the present paper will hopefully shed some light concerns the relative importance of interband and intraband information in the context of different experiments. The question[4,36] has been raised whether in coherent optical experiments interband densities such as $Y=\langle \hat{d}\hat{c}\rangle$ and intraband densities such as $C=\langle \hat{c}^\dagger \hat{c}\rangle$ or $D=\langle \hat{d}^\dagger \hat{d}\rangle$ contain the same or complementary information about the system under investigation. For example, the comparison of FWM data with tetrahertz-emission signals[37] and transmissive electrooptic sampling data[38] indicates manifest differences between coherent beats involving interband or intraband polarizations. In contrast, two-pulse tetrahertz-emission spectroscopy[39] proves that intraband polarizations produced by the first pulse are sensitive to the optical phase of the second, revealing a strong interrelation between intraband and interband polarizations.[40] A central result of the present paper, independent of a specific order in the optical field, may be expressed as follows: In the coherent dynamics generated by the Hamiltonian (2.2) the set of interband functions exhaustively characterizes the evolution of the system. All other functions of intraband or mixed type can be eliminated by virtue of the contraction and factorization rules. The number of interband functions being involved is determined by the order in the driving field.

There are of course a number of open questions not discussed in the present paper. In order to stimulate further work let us mention here at least two examples.

(i) Our analysis so far has been restricted to the coherent dynamics generated by the relevant Hamiltonian. It would be desirable to supplement this kind of dynamics with some dissipative terms having their origin either in the influence of a stochastic environment or in the neglect of higher correlations. An interesting problem in this context is the sensitivity of our rigorous theorems to dissipative interactions. We expect that the factorization theorem is the first one that will be violated since it has to do with the sensitive phases. The factorization of densities into transition amplitudes is a manifestation of the coherence of the excitation that will be destroyed in a dissipative situation. The intraband densities as well as densities of mixed type such as $Z=\langle \hat{c}^\dagger \hat{d}^\dagger \hat{d}\hat{c}\hat{d}\hat{c}\rangle$ then become independent variables.

(ii) An extension of the theory that must be considered sooner or later is the inclusion of phonon dynamics into the hierarchy, which has been, up to now, purely electronic. This kind of extension would also provide a starting point for a proper treatment of an important class of dissipative effects.

ACKNOWLEDGMENTS

Helpful discussions with G. Bartels and H. G. Roskos are gratefully acknowledged.

APPENDIX A: PROOF OF THE EXPANSION THEOREM

The proof of the expansion theorem uses complete induction in n. (i) Using (2.3) and (4.3), Theorem 1 is valid for $n=0$:

$$\hat{n}_{c/v}|t\rangle^{(0)}=e^{-(i/\hbar)H_0 t}\hat{n}_{c/v}|0\rangle=0|0\rangle \ .$$

(ii) Assuming the theorem to be valid for n and using (4.2), $|t\rangle^{(n+1)}$ can be decomposed

$$|t\rangle^{(n+1)}=\sum_{n_p=n,n-2,\ldots\geq 0}(|n_p,n_p,n,t\rangle^{(+)} +|n_p,n_p,n,t\rangle^{(-)})$$

with

$$|n_p,n_p,n,t\rangle^{(\pm)}:=\frac{1}{i\hbar}\int_{-\infty}^{t}dt' e^{-(i/\hbar)H_0(t-t')} \times H_{EP}^{(\pm)}(t')|n_p,n_p,n,t'\rangle \ .$$

Using (2.3) we see that

$$\hat{n}_{c/v}|n_p,n_p,n,t\rangle^{(\pm)}=(n_p\pm 1)|n_p,n_p,n,t\rangle^{(\pm)} \ .$$

According to the induction hypothesis, the inequality $n_p\leq n$ holds and $n-n_p$ is even. Thus we conclude that $n_p\pm 1\leq n+1$ and $(n+1)-(n_p\pm 1)$ is even. Therefore the theorem holds for $n+1$ and, combining (i) and (ii), for all n.

APPENDIX B: PROOF OF THE TRUNCATION THEOREM

According to the expansion theorem the expectation value $X(m,n_g,n_u)$ can be expanded with respect to the optical field:

$$X(t)=\sum_{n=0}^{\infty}X^{(n)}(t) \quad \text{with } X^{(n)}\propto E^N \ ,$$

$$X^{(n)}(t)=\sum_{k=0}^{n}\sum_{n_p}\left\langle n_p,n_p,n-k,t\left|\hat{c}^{\dagger}[n_u]_{\lambda}\hat{Y}^{\dagger}[n_p^{+}]_{\nu}^{\nu'}\right.\right. \times\sum_{n_p'}\hat{Y}[n_p^{-}]_{\mu}^{\mu'}\hat{c}[n_u]_{\kappa}\left|\left.n_p',n_p',k,t\right\rangle\right.$$

with $n_p=n-k-2p$; $n_p'=k-2p'$; $p,p'\geq 0$; and $m=n_p^{+}+n_p^{-}+2|n_u|$. For positive (negative) n_u, the operator $\hat{Y}[n_p^{-}]\hat{c}[n_u]$ tries to annihilate $n_p^{-}+|n_u|$ electrons (holes) from $|n_p',n_p',k,t\rangle$. This gives a nonvanishing result only for $n_p'\geq n_p^{-}+|n_u|$ and results in a state with $n_p'-n_p^{-}-|n_u|$ electrons (holes). Similarly the creators acting to the left yield zero, except for $n_p\geq n_p^{+}+|n_u|$. The resulting states contain $n_p-n_p^{+}-|n_u|$ electrons (holes). The scalar product of bra and ket requires $n_p-n_p^{+}-|n_u|=n_p'-n_p^{-}-|n_u|:=n_s\geq 0$. Summation of these two equalities yields $2n_s=n_p+n_p'-2|n_u|-n_p^{+}-n_p^{-}=n-2p-2p'-m$. Thus

$$n=m+2(p+p'+n_s) \quad \text{with } p+p'+n_s\geq 0$$

holds for all nonvanishing $X^{(n)}$.

APPENDIX C: PROOF OF THE CONTRACTION THEOREM

Let $\hat{I}_l^{n_u}$ for positive (negative) n_u denote an operator with the following properties. All states with definite particle numbers are eigenstates of $\hat{I}_l^{n_u}$. In the subspace of all Fock states with $n_s\leq l$ electrons (holes) and $n_s+|n_u|$ holes (electrons), $\hat{I}_l^{n_u}$ acts like the identity. With this definition, in a first step, we prove

$$\langle\hat{c}^{\dagger}[n_u]_{\lambda}\hat{Y}^{\dagger}[n_p^{+}]_{\nu}^{\nu'}(\hat{1}-\hat{I}_l^{n_u})\hat{Y}[n_p^{-}]_{\mu}^{\mu'}\hat{c}[n_u]_{\kappa}\rangle =O(E^{m+2l+2}) \ . \quad \text{(C1)}$$

The proof is parallel to Appendix B and we keep the notations used there. Inserting the expansion of the Schrödinger state according to the expansion theorem, we realize that $(\hat{1}-\hat{I}_l^{n_u})$ in (C1) only acts on states that contain $n_s=n_p-n_p^{+}-|n_u|=n_p'-n_p^{-}-|n_u|$ electrons (holes) and n_s+m holes (electrons). On account of the identity property of $\hat{I}_l^{n_u}$, nonvanishing contributions require $n_s=l+l'$ with $l'\geq 1$. Thus the expectation value is of order

$$n=m+2(p+p'+n_s)=m+2l+2(p+p'+l') \ ,$$

with $p+p'+l'\geq 1$, as stated by (C1).

For positive/negative n_u, a possible choice for $\hat{I}_l^{n_u}$ is

$$\hat{I}_l^{n_u}=\sum_{k=0}^{l}g_{|n_u|,k}\prod_{p=0}^{k-1}(\hat{n}_{c/v}-p)\prod_{p'=0}^{k+|n_u|-1}(\hat{n}_{v/c}-p') \ .$$

Splitting the sum in $k=l$ and $k=0,\ldots,l-1$, the proof of the required properties of $\hat{I}_l^{n_u}$ is straightforward, using complete induction with respect to l. The contraction theorem follows from (C1), inserting the above choice of $\hat{I}_l^{n_u}$ and the transcription

$$\prod_{p=0}^{k-1}(\hat{n}_{c/v}-p)=\sum_{(i)_k}\hat{c}^{\dagger}[\pm k]_{(i)_k}\hat{c}[\pm k]_{(i)_k} \ , \quad \text{(C2)}$$

which again can be shown by induction.

APPENDIX D: PROOF OF THE FACTORIZATION THEOREM

In a first step, we calculate a contribution $\langle\hat{Y}^{\dagger}[n_p^{+}]_{\nu}^{\nu'}\hat{P}_l\hat{Y}[n_p^{-}]_{\mu}^{\mu'}\rangle^{(n_p^{+}+n_p^{-}+p)}$ with $p\leq 2l$, using the

expansion theorem as we did in Appendix B. Using the same notations as in Appendix B ($n \equiv n_p^+ + n_p^- + p$), we only have to know the action of $\hat{P}_l$ on states with $n_s = n_p' - n_p^- = n_p - n_p^+$ electrons and holes. From the expansion theorem we know that the inequalities $n_p \le n_p^+ + n_p^- + p - k$ and $n_p' \le k$ hold, thus $2n_s = n_p - n_p^+ + n_p' - n_p^- \le p \le 2l$. On these states, $\hat{P}_l$ acts like the projector $|0\rangle\langle 0|$ and gives a nonvanishing result only for $n_s = 0$. Therefore the only contributions to the expectation value are those with $n_p = n_p^+$, $n_p' = n_p^-$, and $n_p^- \le k \le n_p^- + p$. Substituting $k' := k - n_p^-$ yields

$$\langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{P}_l \hat{Y}[n_p^-]_\mu^{\mu'} \rangle^{(n_p^+ + n_p^- + p)} = \sum_{k'=0}^{p} \langle n_p^+, n_p^+, n_p^+ + p - k', t | \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} | 0 \rangle \times \langle 0 | \hat{Y}[n_p^-]_\mu^{\mu'} | n_p^-, n_p^-, n_p^- + k', t \rangle \ . \quad \text{(D1)}$$

In a second step we decompose the left-hand side and the right-hand side of the factorization theorem separately into contributions proportional to $E^{n_p^+ + n_p^- + p}$. Using (D1), we obtain

$$(\langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{P}_l \hat{Y}[n_p^-]_\mu^{\mu'} \rangle \langle \hat{P}_l \rangle)^{(n_p^+ + n_p^- + p)} = \sum_{k=0}^{p} \langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{P}_l \hat{Y}[n_p^-]_\mu^{\mu'} \rangle^{(n_p^+ + n_p^- + p - k)} \langle \hat{P}_l \rangle^{(k)}$$

$$= \sum_{k=0}^{p} \sum_{k'=0}^{p-k} \sum_{k''=0}^{k} S(p-k-k', k', k-k'', k'') \ .$$

$$(\langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{P}_l \rangle \langle \hat{P}_l \hat{Y}[n_p^-]_\mu^{\mu'} \rangle)^{(p + n_p^+ + n_p^-)} = \sum_{k=0}^{p} \langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{P}_l \rangle^{(n_p^+ + k)} \langle \hat{P}_l \hat{Y}[n_p^-]_\mu^{\mu'} \rangle^{(n_p^- + p - k)}$$

$$= \sum_{k=0}^{p} \sum_{k''=0}^{k} \sum_{k'=0}^{p-k} S(k-k'', k', p-k-k', k'')$$

with

$$S(n_1, n_2, n_3, n_4) := \langle n_p^+, n_p^+, n_p^+ + n_1, t | \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} | 0 \rangle \times \langle 0 | \hat{Y}[n_p^-]_\mu^{\mu'} | n_p^-, n_p^-, n_p^- + n_2, t \rangle \times \langle 0,0,n_3,t | 0 \rangle \langle 0 | 0,0,n_4,t \rangle \ .$$

Rearranging the order of summations such that $\sum_k$ becomes the innermost and substituting $\tilde{k} := p - k - k' + k''$ yields

$$(\langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{P}_l \rangle \langle \hat{P}_l \hat{Y}[n_p^-]_\mu^{\mu'} \rangle)^{(n_p^+ + n_p^- + p)} = \sum_{k'=0}^{p} \sum_{k''=0}^{p-k'} \sum_{k=k''}^{p-k'} S(k-k'', k', p-k-k', k'')$$

$$= \sum_{k'=0}^{p} \sum_{k''=0}^{p-k'} \sum_{\tilde{k}=k''}^{p-k'} S(p-\tilde{k}-k', k', \tilde{k}-k'', k'')$$

$$= \sum_{\tilde{k}=0}^{p} \sum_{k'=0}^{p-\tilde{k}} \sum_{k''=0}^{p-\tilde{k}} S(p-\tilde{k}-k', k', \tilde{k}-k'', k'')$$

$$= (\langle \hat{Y}^\dagger[n_p^+]_\nu^{\nu'} \hat{P}_l \hat{Y}[n_p^-]_\mu^{\mu'} \rangle \langle \hat{P}_l \rangle)^{(n_p^+ + n_p^- + p)} \ .$$

Thus the factorization theorem is valid to all orders $O(E^{n_p^+ + n_p^- + p})$ with $p \le 2l$. According to the truncation theorem, the first derivations will be of order $O(E^{n_p^+ + n_p^- + 2l + 2})$.

The projector properties of $\hat{P}_l$ can be proven if we substitute (C2) into the definition and obtain $\hat{P}_l$ in a form that is not normally ordered:

$$\hat{P}_l = \hat{1} + \sum_{k=1}^{l} p_k \sum_{k'=0}^{k-1} (\hat{n}_e - k')(\hat{n}_h - k') \ .$$

Written in this form, $\hat{P}_l |p\rangle$, with $|p\rangle$ denoting a p-pair Schrödinger state, can easily be shown to be zero for $1 \le p \le l$ and $|0\rangle$ for $p = 0$, by splitting the sum into $k = 1, \ldots p$, $k = p$, and $k \ge p$. Furthermore, all states with a definite particle number are obviously eigenstates of $\hat{P}_l$, as required for the factorization theorem.

APPENDIX E: PROOF OF THEOREM 6

It remains to prove that the calculation of a variable $X(m, n_g, n_u)$ with $m + \max(|n_g| - 1, 0) \le n$ to orders $n - \max(|n_g| - 1, 0)$ with the help of the contraction and factorization theorems does not involve density matrices $X'(m', n_g', n_u')$ with $m' + \max(|n_g'| - 1, 0) > n$ or accuracies of X' higher than $n - \max(|n_g'| - 1, 0)$.

The use of the contraction theorem requires the knowledge of variables $X'(m', n_g, 0)$ with $2k = m' - m \le 2l = [n - \max(|n_g| - 1, 0)] - m$ and thus $m' + \max(|n_g| - 1, 0) \le n$ too. X' has the same gap order as X and has to be calculated to the same accuracy.

The evaluation of $X(m, n_g, 0) = \langle Y^\dagger[n_p^+] Y[n_p^-] \rangle$ with the help of the factorization theorem exact to orders $n - \max(|n_g| - 1, 0)$ according to Eq. (4.13) involves transition densities $X'(m', n_g', 0) = \langle Y[k] \rangle$ with

$k \le \max(n_p^+, n_p^-) + l$, where l is defined by $n_p^+ + n_p^- + 2l = n - \max(|n_g| - 1, 0)$. For these transition densities, $m' = n_g' = k \ge 1$ holds and thus $m' + \max(|n_g'| - 1, 0) = 2k - 1 \le 2\max(n_p^+, n_p^-) + 2l - 1 \le n$.

In contrast to the contraction theorem, the factorization theorem involves variables X' with $n_g' = k \neq n_g$. Therefore we have to verify that the calculation of X to $[n - \max(|n_g| - 1, 0)]$th order does not require X' to be known to orders higher than $n - \max(|n_g'| - 1, 0) = n - k + 1$. According to (4.14), X' occurs only in products of the form $\prod \langle Y[k^+] \rangle^* \prod \langle Y[k^-] \rangle$ with $\sum k^- \sum k^+ = n_g$. This product has to be evaluated to order $n - \max(|n_g| - 1, 0)$. One of the factors is X'; the product of the other factors is at least of order $||n_g| - k|$. Thus X' has to be calculated to orders $[n - \max(|n_g| - 1, 0)] - ||n_g| - k| \le n - k + 1$.

We conclude that within a $\chi^{(n)}$ calculation the factorization theorem as well as the contraction theorem can be applied without enlargement of the set of relevant density matrices or the required accuracy.

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