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Dynamics of excitations in molecular condensates under the influence of an ultrashort pulse

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Abstract. A new method for the description of simultaneous creation, annihilation, propagation and relaxation of excitations in exciton (electron) systems interacting with a phonon bath and an ultrashort quantum optical pulse is developed on the basis of generalized master equations derived by using a time-dependent projector. It is shown, that standard theories starting from an already created excitation at a given place, without any off-diagonal elements of the exciton density matrix, have a very limited area of application because off-diagonal elements are generally nonzero once the excited pulse switches off. Derived equations also describe the influence of quantum statistics of the pulse.

1. Introduction

Investigation of the interaction of electromagnetic fields with quantum systems gets more important all the time due to the rapid development of experimental techniques. In particular, ultrafast optical spectroscopy, which takes advantages of optical pulses of several tens of femtosecond duration, is developing fast [1]. New experiments make it necessary to develop theories which are more powerful in the description of the interaction of electromagnetic fields with matter than the classical optical Bloch equations.

Optical Bloch equations describe the interaction of a two-level atom with a monochromatic classical wave and the interaction of the two-level atom with a bath is described phenomenologically using the longitudinal and transversal relaxation times [2]. The inclusion of the interaction with a bath on a microscopic level leads to the generalized optical Bloch equations [3]. They contain in general three relaxation times, which are field dependent. Their use is inevitable in the case of strong fields. They reduce to the classical Bloch equations in the limit of weak fields. The Bloch equations can also be generalized for atoms which have more than two levels and for fields composed of two or more monochromatic components [2].

Great attention has been devoted to the description of the interaction of ultrashort classical pulses with excitations in semiconductors during last years [4]. The spacial periodicity of studied materials enables us to apply conveniently the description based on Green function techniques. The method of equations of motion for reduced density matrices has been developed and successfully applied [5, 6] when studying Coulomb, exchange and electron (exciton)–phonon interactions in semiconductors. The other method used in this

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field is the method of nonequilibrium Green functions. It can also be successfully applied when static disorder of material is present [7].

In the case of excitations in various molecular condensates interacting with a phonon bath the above mentioned approaches are not suitable. The reason is a combination of the strength of the static disorder as compared with the band width and the strength of the on-site coupling to phonons. Description based on (generalized) master equations (see e.g. [8–10]) is standardly used in this field. However, an initially created excitation is always supposed in standard theories, and master equations then describe only the dynamics of an excitation interacting with a phonon bath. On the other hand, with respect to the progress of experimental techniques, it is important to understand also the initial period of the time development of excitation, i.e. the period in which the excitation is being created but in which it is already influenced by propagation and relaxation.

We develop a new method which allows us to treat simultaneous creation, relaxation and propagation of an excitation in molecular condensates interacting with ultrashort pulses [11]. The method is based on the use of the projection superoperator technique based on a time-dependent projector and it provides a generalized master equation for the reduced density matrix of the exciton subsystem. It represents a generalization of standard approaches based on time-convolutionless master equations [12–14]. Similar to the standard approaches, the perturbation approximation in both the exciton–photon and the exciton–phonon coupling constants is invoked in a rather sophisticated manner.

After the pulse is gone, the method provides the same equations for the excitation dynamics as were derived by the standard approaches which include off-diagonal matrix elements of the exciton-reduced density matrix. However, the method shows that after the pulse switches off, off-diagonal matrix elements are in general nonzero. This means that the standard approaches have a very limited area of application because they suppose an initially created excitation at a given place and they assume zero initial off-diagonal elements of the exciton reduced density matrix.

The method also substantially differs from all the above-mentioned theories in the following. It is capable to describe effects connected with statistical properties of optical fields including nonclassical, i.e. purely quantum, properties like squeezing of vacuum fluctuations. This makes the method promising for applications in this direction, because all the above-mentioned methods describe only the interaction of matter with classical deterministic fields.

The paper is divided as follows. Section 2 describes a model of the system under investigation composed of exciton, phonon and photon subsystems in the mutual interaction. Section 3 is devoted to generalized master equations for coupled systems. Section 4 contains a derivation of equations for the exciton-reduced density matrix. Comparison of equations derived in section 4 with those from standard theories is given in section 5. Section 6 summarizes the obtained results and gives conclusions.

2. Description of a model

Our system consists of exciton (or electron), photon and phonon subsystems with the exciton–photon and exciton–phonon interactions. Hamiltonians of the free exciton (\hat{H}_e),

photon (\hat{H}_f) and phonon (\hat{H}_{ph}) subsystems are given as follows

$$\begin{aligned}\hat{H}_e &= \sum_{m,n} J_{mn} \hat{c}_m^\dagger \hat{c}_n \\ \hat{H}_f &= \sum_K \hbar \omega_K \hat{a}_K^\dagger \hat{a}_K \\ \hat{H}_{ph} &= \sum_k \hbar \Omega_k \hat{b}_k^\dagger \hat{b}_k\end{aligned}\quad (1)$$

where \hat{c}_m^\dagger (\hat{c}_m) means the creation (annihilation) operator of exciton at the m th site of the lattice (Frenkel excitons are considered), \hat{a}_K^\dagger (\hat{a}_K) represents the creation (annihilation) operator of the K th photon mode and \hat{b}_k^\dagger (\hat{b}_k) is the creation (annihilation) operator of the k th phonon mode. The coefficients J_{mn} describe energies of the free exciton subsystem for $m = n$ and transfer in the exciton subsystem given by overlaps of wavefunctions for $m \neq n$. The excitonless state $|0\rangle$ is supposed to have zero energy. The symbol ω_K (Ω_k) stands for the frequency of the K th (k th) mode of photon (phonon) field. The symbol \sum_K (\sum_k) means summation over all photon (phonon) modes and \sum_m denotes summation over all exciton states; \hbar is the reduced Planck constant.

The exciton operators \hat{c}_m^\dagger and \hat{c}_m obey the Pauli commutation relations, whereas the photon and phonon operators \hat{a}_K^\dagger , \hat{a}_K , \hat{b}_k^\dagger and \hat{b}_k obey the boson commutation relations. With respect to the restriction of the following calculations to the excitonless state and to states with only one exciton, we can also use the obtained results without any change for an electron subsystem instead of the exciton one, regardless of the fact that electron operators are of the fermion type.

The exciton–photon interaction Hamiltonian \hat{H}_{e-f} in the rotating wave approximation reads [15]

$$\hat{H}_{e-f} = \sum_{m,K} \hbar \omega_{K_0} F_K^m (\hat{a}_K \hat{c}_m^\dagger + \hat{a}_{-K}^\dagger \hat{c}_m) \quad (2)$$

where ω_{K_0} is a typical photon frequency. The coupling constants F_K^m must fulfil the relations $F_K^m = F_{-K}^{m*}$ (* denotes complex conjugation) in order to ensure hermiticity of \hat{H}_{e-f} . The exciton–photon coupling constants F_K^m are given in the dipole approximation by

$$F_K^m = -\frac{1}{\hbar \omega_{K_0}} \sqrt{\frac{\hbar}{2\epsilon_0 V \omega_K}} \frac{e}{m_e} \epsilon_K \cdot \langle m | \hat{\mathbf{p}} | 0 \rangle \exp(i\mathbf{K} \cdot \mathbf{r}_m). \quad (3)$$

Here e ($e < 0$) is the charge of electron, m_e the mass of electron, $\hat{\mathbf{p}}$ the momentum operator of the electron, \mathbf{r}_m the mean position of the m th atom in a lattice, $|0\rangle$ describes the excitonless state, $|m\rangle$ the state with one exciton localized at the m th site of the lattice and the dot means the scalar product; ϵ_K is the polarization vector of the K th mode of the photon field, ϵ_0 permittivity of a vacuum and V the quantization volume of the electromagnetic field. We supposed that $\epsilon_K = \epsilon_{-K}^*$ and the coefficients $\langle m | \hat{\mathbf{p}} | 0 \rangle$ to be real in expression (2) for \hat{H}_{e-f} .

The exciton–phonon interaction is described by the interaction Hamiltonian \hat{H}_{e-ph} in the form [15, 10]

$$\hat{H}_{e-ph} = \frac{1}{\sqrt{N}} \sum_{m,k} \hbar \Omega_k G_k^m \hat{c}_m^\dagger \hat{c}_m (\hat{b}_k + \hat{b}_{-k}^\dagger) \quad (4)$$

with only the site-diagonal exciton–phonon coupling included. Hermiticity of \hat{H}_{e-ph} leads to the restriction $G_k^m = G_{-k}^{m*}$ for the exciton–phonon coupling constants. The dependence

of the exciton–phonon coupling constants G_k^m on the site index m and the mode index k is determined according to the type of phonons (optical or acoustic) and according to the model of the exciton–phonon interaction. N means the number of phonon modes. Hamiltonian (4) describes deformation of the lattice around a given site m after it was occupied by an exciton (polaron effect). Terms describing the site-off-diagonal exciton–phonon coupling, which describe the motion of excitons promoted by phonons, are missing here, because we suppose that they are negligible (for a detailed discussion see [10]).

Finally we introduce the Liouville superoperators

$$\mathcal{L}_\alpha = \frac{1}{\hbar}[\hat{H}_\alpha,] \quad (5)$$

for all Hamiltonians \hat{H}_α defined above, i.e. for $\alpha = e, f, \text{ph}, e\text{-}f, e\text{-}\text{ph}$. We also introduce the Liouville superoperators for the exciton subsystem \mathcal{L}_S ($\mathcal{L}_S = \mathcal{L}_e$), compound photon and phonon subsystem \mathcal{L}_R ($\mathcal{L}_R = \mathcal{L}_f + \mathcal{L}_{\text{ph}}$), whole system without interaction \mathcal{L}_0 ($\mathcal{L}_0 = \mathcal{L}_e + \mathcal{L}_f + \mathcal{L}_{\text{ph}}$), for the interaction part of the whole system \mathcal{L}_{int} ($\mathcal{L}_{\text{int}} = \mathcal{L}_{e\text{-}f} + \mathcal{L}_{e\text{-}\text{ph}}$) and for the whole system \mathcal{L} ($\mathcal{L} = \mathcal{L}_e + \mathcal{L}_f + \mathcal{L}_{\text{ph}} + \mathcal{L}_{e\text{-}f} + \mathcal{L}_{e\text{-}\text{ph}}$).

3. Projection superoperator formalism

A projection superoperator formalism forms the basis for the method. We have a system which consists of three subsystems, but we orientate our attention mainly to the exciton one. So our aim is to eliminate the photon and phonon subsystems and to obtain equations which govern the dynamics of the exciton subsystem only. Such a task can be solved by the use of projection superoperator techniques applied to the Liouville equation for the density matrix of the composite system [16, 17]. The projection superoperator techniques based on time-independent projectors [18–21] are used by standard methods for the description of exciton interaction with an inertial phonon reservoir [12–14]. However, the situation is more complex in our case, because the photon subsystem with a density matrix which is strongly time-dependent (it describes an ultrashort optical pulse) should also be projected off. A strong time dependence of the photon density matrix does not exclude, in principle, the application of time-independent projectors, but their use is not convenient. The application of a time-dependent projector in such cases is natural and convenient.

It was revealed when studying exciton–phonon systems, that the time-convolutionless generalized master equations are much more suitable for dealing with such systems, because they are able to describe correctly the relaxation of excitons to the equilibrium state for long times in the finite (second) order of the perturbation theory in the exciton–phonon coupling constants. The time-convolution generalized master equations do not have this property unless other (Markovian) approximations are involved [22].

For the above reason, we use the so-called time-convolutionless formalism. This name comes from the fact that it yields time-convolutionless (i.e. time-local) equations upon application of the time-independent projectors. In our case, owing to the time-dependent form of the projector [23], the equations are also time-nonlocal. Anyway, technical advantages remain. Our approach is based on the master equations for coupled systems derived in [24] (for an application see [25]) for a system (described by the density matrix $\hat{\rho}(t)$) composed of a small system described by $\hat{\rho}_S(t)$ ($\hat{\rho}_S(t) = \text{Tr}_R\{\hat{\rho}(t)\}$ Tr_R is the trace over reservoir) and reservoir described by $\hat{\rho}_R(t)$ ($\hat{\rho}_R(t) = \text{Tr}_S\{\hat{\rho}(t)\}$ Tr_S represents the trace over the small system). The derivation of the master equations is based on the application of the time-dependent projector $\mathcal{P}(t)$ of the form

$$\mathcal{P}(t)\hat{X} = \hat{\rho}_S(t) \text{Tr}_S\{\hat{X}\} + \hat{\rho}_R(t) \text{Tr}_R\{\hat{X}\} - \hat{\rho}_S(t)\hat{\rho}_R(t) \text{Tr}\{\hat{X}\} \quad (6)$$

where $\text{Tr} = \text{Tr}_S \text{Tr}_R$ denotes the trace over the whole system and \hat{X} stands for an arbitrary operator.

The standard procedure for the derivation of the time-convolutionless generalized master equations leads to the following master equation

$$\begin{aligned} \frac{d}{dt} \mathcal{P}(t) \hat{\rho}(t) = & -i\mathcal{P}(t) \mathcal{L} \mathcal{P}(t) \hat{\rho}(t) - i\mathcal{P}(t) \mathcal{L}_{\text{int}} [\theta(t) - 1] \mathcal{P}(t) \hat{\rho}(t) \\ & - i\mathcal{P}(t) \mathcal{L}_{\text{int}} \theta(t) G(t, t_0) \mathcal{D}(t_0) \hat{\rho}(t_0) \end{aligned} \quad (7)$$

where

$$\theta(t) - 1 = \frac{\epsilon(t)}{1 - \epsilon(t)} = \epsilon(t) + \epsilon^2(t) + \dots \quad (8)$$

$$\epsilon(t) = -i \int_{t_0}^t ds G(t, s) \mathcal{D}(s) \mathcal{L}_{\text{int}} \mathcal{P}(s) G(t - s) \quad (9)$$

$$G(t, s) = \exp_{\leftarrow} \left[-i \int_s^t d\tau \mathcal{D}(\tau) \mathcal{L} \right] \quad (10)$$

$$G(t - s) = \exp[i\mathcal{L}(t - s)]. \quad (11)$$

In (7)–(11), $\mathcal{D}(t) = 1 - \mathcal{P}(t)$ is projector and the symbol \exp_{\leftarrow} denotes a time-ordered exponential with time arguments increasing to the left. The initial condition term (the last term in (7)) is zero when the factorized initial density matrix $\hat{\rho}(t_0) = \hat{\rho}_S(t_0) \hat{\rho}_R(t_0)$ is supposed (then $\mathcal{D}(t_0) \hat{\rho}(t_0) = 0$). The equations for the reduced density matrices $\hat{\rho}_S(t)$ and $\hat{\rho}_R(t)$ are derived from (7) by the application of the operations Tr_R and Tr_S , respectively.

4. Equations for the exciton-reduced density matrix

We derive equations for matrix elements of the exciton-reduced density matrix $\hat{\rho}_e(t)$ on the basis of the master equation (7). First we divide our three-component system into a small system and a reservoir. The exciton subsystem constitutes the small system ($\hat{\rho}_S(t) = \hat{\rho}_e(t)$) and the photon and phonon subsystems form together the reservoir ($\hat{\rho}_f(t) = \text{Tr}_{\text{ph}}\{\hat{\rho}_R(t)\}$, $\hat{\rho}_{\text{ph}}(t) = \text{Tr}_f\{\hat{\rho}_R(t)\}$, where Tr_{ph} and Tr_f mean the traces over the phonon and the photon subsystem, respectively). We suppose that the exciton and photon–phonon subsystems are statistically independent at the beginning of interaction at time t_0 , so the initial density matrix $\hat{\rho}(t_0)$ factorizes ($\hat{\rho}(t_0) = \hat{\rho}_S(t_0) \hat{\rho}_R(t_0)$) and the initial condition term in (7) is zero. We further restrict ourselves to the second order of the perturbation approximation in the interaction Liouville superoperator \mathcal{L}_{int} .

The master equation under these constraints reads

$$\begin{aligned} \frac{d}{dt} \mathcal{P}(t) \hat{\rho}(t) = & -i\mathcal{P}(t) \mathcal{L} \mathcal{P}(t) \hat{\rho}(t) - \mathcal{P}(t) \mathcal{L}_{\text{int}} \int_{t_0}^t ds \exp[-i\mathcal{L}_0(t - s)] [1 - \mathcal{P}(s)] \mathcal{L}_{\text{int}} \mathcal{P}(s) \\ & \times \exp[i\mathcal{L}_0(t - s)] \mathcal{P}(t) \hat{\rho}(t) + o(\mathcal{L}_{\text{int}}^2). \end{aligned} \quad (12)$$

The relation $\mathcal{P}(t) \mathcal{L}_0 \mathcal{D}(t) = 0$ was used when deriving equation (12). The application of Tr_R and Tr_S to the master equation in (12) provides a coupled set of equations for the reduced density matrices $\hat{\rho}_S(t)$ and $\hat{\rho}_R(t)$ in the form

$$\begin{aligned} \frac{d}{dt} \hat{\rho}_S(t) = & -i\mathcal{L}_S \hat{\rho}_S(t) - i \text{Tr}_R \{ \mathcal{L}_{\text{int}} \hat{\rho}_R(t) \} \hat{\rho}_S(t) - \text{Tr}_R \left\{ \mathcal{L}_{\text{int}} \int_{t_0}^t d\tau \exp[-i\mathcal{L}_0(t - \tau)] \right. \\ & \left. \times [1 - \mathcal{P}(\tau)] \mathcal{L}_{\text{int}} \mathcal{P}(\tau) \exp[i\mathcal{L}_0(t - \tau)] \hat{\rho}_R(\tau) \right\} \hat{\rho}_S(t) + o(\mathcal{L}_{\text{int}}^2) \end{aligned} \quad (13)$$

$$\begin{aligned} \frac{d}{dt} \hat{\rho}_R(t) = & -i\mathcal{L}_R \hat{\rho}_R(t) - i \text{Tr}_S \{ \mathcal{L}_{\text{int}} \hat{\rho}_S(t) \} \hat{\rho}_R(t) - \text{Tr}_S \left\{ \mathcal{L}_{\text{int}} \int_{t_0}^t d\tau \exp[-i\mathcal{L}_0(t-\tau)] \right. \\ & \left. \times [1 - \mathcal{P}(\tau)] \mathcal{L}_{\text{int}} \mathcal{P}(\tau) \exp[i\mathcal{L}_0(t-\tau)] \hat{\rho}_S(t) \right\} \hat{\rho}_R(t) + o(\mathcal{L}_{\text{int}}^2). \end{aligned} \quad (14)$$

We suppose that the influence of the exciton subsystem on the photon–phonon subsystem (reservoir) is negligible. This leads to a strong simplification of equation (14) for $\hat{\rho}_R(t)$ in which only the first term on the right-hand side remains.

The photon and phonon subsystems are statistically independent at the beginning of interaction and, neglecting the influence of exciton subsystem on the photon and phonon ones, no coupling between the photon and phonon subsystems can develop during the interaction. The density matrix of reservoir thus factorizes for all times, $\hat{\rho}_R(t) = \hat{\rho}_f(t) \hat{\rho}_{\text{ph}}(t)$, and the density matrices $\hat{\rho}_f(t)$ and $\hat{\rho}_{\text{ph}}(t)$ of the photon and the phonon subsystem, respectively, obey the following equations,

$$\frac{d}{dt} \hat{\rho}_f(t) = -i\mathcal{L}_f \hat{\rho}_f(t) \quad (15)$$

$$\frac{d}{dt} \hat{\rho}_{\text{ph}}(t) = -i\mathcal{L}_{\text{ph}} \hat{\rho}_{\text{ph}}(t). \quad (16)$$

The phonon subsystem is supposed to be in the thermal equilibrium and it is described by the canonical density matrix $\hat{\rho}_{\text{ph}}(t)$,

$$\hat{\rho}_{\text{ph}}(t) = \hat{\rho}_{\text{ph}}(t_0) = \frac{\exp[-\beta \hat{H}_{\text{ph}}]}{\text{Tr}_{\text{ph}} \{ \exp[-\beta \hat{H}_{\text{ph}}] \}} \quad (17)$$

$\beta = 1/(k_B T)$, k_B being the Boltzmann constant and T the absolute temperature.

The photon subsystem is described in general by a density matrix $\hat{\rho}_f(t)$ obeying equation (15) [26, 27].

After prescribing the time development of $\hat{\rho}_R(t)$ driven by equations (15) and (16) and inserting $\hat{\rho}_R(t)$ into (13), equation (13) then represents a closed equation for the exciton-reduced density matrix $\hat{\rho}_e(t)$ in the form,

$$\begin{aligned} \frac{d}{dt} \hat{\rho}_e(t) = & -i\mathcal{L}_e \hat{\rho}_e(t) - i \text{Tr}_R \{ \mathcal{L}_{\text{int}} \hat{\rho}_R(t) \} \hat{\rho}_e(t) - \text{Tr}_R \left\{ \mathcal{L}_{\text{int}} \int_{t_0}^t d\tau \exp[-i\mathcal{L}_0(t-\tau)] \right. \\ & \left. \times [1 - \mathcal{P}(\tau)] \mathcal{L}_{\text{int}} \exp[i\mathcal{L}_0(t-\tau)] \hat{\rho}_R(t) \right\} \hat{\rho}_e(t) + o(\mathcal{L}_{\text{int}}^2). \end{aligned} \quad (18)$$

The resulting equation for $\hat{\rho}_e(t)$ is nonlinear and integro-differential. This is connected with the special type of the time-dependent projector (6) which was used in its derivation.

We make a note about the origin of nonlinearity in the equation for $\hat{\rho}_e(t)$ derived above. The application of the projector (6) without any approximation leads to the set of two coupled nonlinear integro-differential equations for the reduced density matrices $\hat{\rho}_e(t)$ for a small system and $\hat{\rho}_R(t)$ for a reservoir. The nonlinearity of the obtained equations is a consequence of the fact that we remove from the description the part of the whole density matrix (given by the expression $\hat{\rho}(t) - \hat{\rho}_e(t) \hat{\rho}_R(t)$) which describes correlations between two coupled systems originating from their mutual interaction. Thus nonlinear terms in the equations for the reduced density matrices $\hat{\rho}_e(t)$ and $\hat{\rho}_R(t)$ describe mutual correlations between two mutually interacting systems. In order to get rid of nonlinear terms we must limit the accuracy of the description of the mutual interaction of two systems in general. This can be reached by supposing that time-dependent ‘coefficients’ in the resulting equations for $\hat{\rho}_e(t)$ and $\hat{\rho}_R(t)$ (containing implicitly $\hat{\rho}_e(\tau)$ and $\hat{\rho}_R(\tau)$ in various times τ in projectors) can

be well approximated by their forms obtained by the restriction to several lowest orders of their perturbation expansions. The reduced density matrices $\hat{\rho}_e(\tau)$ and $\hat{\rho}_R(\tau)$ in the projector $\mathcal{P}(\tau)$ are then replaced by their forms valid to a given power of the interaction constants and the nonlinearity is lost.

We derive equations for matrix elements of the reduced density matrix $\hat{\rho}_e(t)$ under the restriction that $\hat{\rho}_e(t)$ lies within the space corresponding to the Hilbert space spanned by the excitonless state $|0\rangle$ and by states with one exciton $|m\rangle$. So it can be expressed in the form

$$\hat{\rho}_e(t) = \sum_{m,n} \rho_{mn}^e(t) |m\rangle \langle n| + \sum_m \rho_{m0}^e(t) |m\rangle \langle 0| + \sum_n \rho_{0n}^e(t) |0\rangle \langle n| + \rho_{00}^e(t) |0\rangle \langle 0| \quad (19)$$

where $\rho_{mn}^e(t) = \langle m | \hat{\rho}_e(t) | n \rangle$ and the coefficients ρ_{m0}^e , ρ_{0n}^e and ρ_{00}^e are defined similarly. Doubly and other multiply excited states are disregarded here. This imposes limitations on the strength of the applied pulse which do not, however, seem severe.

When we substitute the expression for $\hat{\rho}_e(t)$ from (19) into equation (18), we obtain equations for the matrix elements $\rho_{mn}^e(t)$, $\rho_{m0}^e(t)$, $\rho_{0n}^e(t)$ and $\rho_{00}^e(t)$. We get in general the system of coupled nonlinear integro-differential equations for $\rho_{mn}^e(t)$, $\rho_{m0}^e(t)$, $\rho_{0n}^e(t)$ and $\rho_{00}^e(t)$. In order to simplify them, we suppose that $\hat{\rho}_e(t)$ in the projector $\mathcal{P}(\tau)$ in (18) is approximated sufficiently by its perturbation expansion limited by several lowest orders in the perturbation parameters in such a way that coefficients in the resulting coupled linear differential equations for the matrix elements of $\hat{\rho}_e(t)$ are precise to the second order in the perturbation parameters. A detailed analysis [11] shows, that it is sufficient to restrict the perturbation expansion for $\hat{\rho}_e(t)$ to the zeroth order of perturbation parameters. Because our goal is to study excitation of the exciton subsystem, we suppose, that the exciton subsystem is in the excitonless state at the beginning of interaction, i.e. $\hat{\rho}_e(t_0) = |0\rangle \langle 0|$. With respect to the form of \hat{H}_e in (1) (state $|0\rangle$ is an eigenstate of \hat{H}_e with zero energy) this state does not evolve in time without inclusion of the exciton-photon interaction, i.e. $\hat{\rho}_e^{(0)}(t) = |0\rangle \langle 0| + O(\text{coupling constants})$.

The method described above provides the following equations for the matrix elements of the exciton-reduced density matrix (details of the derivation can be found in [11])

$$\begin{aligned} \frac{d}{dt} \rho_{mn}^e(t) = & -\frac{i}{\hbar} \sum_p J_{mp} \rho_{pn}^e(t) + \frac{i}{\hbar} \sum_p J_{pn} \rho_{mp}^e(t) - \sum_p G_p^{mn}(t) \rho_{pn}^e(t) - \sum_p G_p^{nm*}(t) \\ & \times \rho_{mp}^e(t) - \sum_p \bar{I}_p^m(t) \rho_{pn}^e(t) - \sum_p \bar{I}_p^{n*}(t) \rho_{mp}^e(t) - i F^m(t) \rho_{0n}^e(t) \\ & + i F^{n*}(t) \rho_{m0}^e(t) + [I_n^m(t) + I_m^{n*}(t)] \rho_{00}^e(t) \end{aligned} \quad (20)$$

$$\begin{aligned} \frac{d}{dt} \rho_{00}^e(t) = & - \sum_l [I_l^{l*}(t) + I_l^l(t)] \rho_{00}^e(t) - i \sum_p F^{p*}(t) \rho_{p0}^e(t) + i \sum_p F^p(t) \rho_{0p}^e(t) \\ & + \sum_{s,p} [\bar{I}_p^{s*}(t) + \bar{I}_s^p(t)] \rho_{sp}^e(t) \end{aligned} \quad (21)$$

$$\begin{aligned} \frac{d}{dt} \rho_{0n}^e(t) = & \frac{i}{\hbar} \sum_p J_{pn} \rho_{0p}^e(t) - \sum_p G_p^{n*}(t) \rho_{0p}^e(t) - \sum_p I_p^{n*}(t) \rho_{0p}^e(t) - \sum_l I_l^{l*}(t) \rho_{0n}^e(t) \\ & + \sum_p [\bar{I}_n^p(t) + \bar{I}_p^n(t)] \rho_{p0}^e(t) - i \sum_p F^{p*}(t) \rho_{pn}^e(t) + i F^{n*}(t) \rho_{00}^e(t) \end{aligned}$$

$$\rho_{m0}^e(t) = \rho_{0m}^{e*}(t). \quad (22)$$

The coefficients J_{mn} describe the inner dynamics of the exciton subsystem, the time-dependent coefficients $G_p^m(t)$ and $G_p^m(t)$ characterize the interaction of the exciton

subsystem with phonons and the time-dependent coefficients $F^m(t)$, $I_p^m(t)$, $\bar{I}_p^m(t)$ and $\tilde{I}_p^m(t)$ emerge from the exciton–photon interaction. The coefficients occurring in equations (20)–(22) are correct to the second power of the exciton–photon and exciton–phonon coupling constants, as will be seen from the ensuing expressions for the coefficients.

The time-dependent coefficients $G_p^{mn}(t)$ and $G_p^m(t)$ having their origin in the exciton–phonon interaction are given as follows

$$G_p^{mn}(t) = \int_0^{t-t_0} d\tau \frac{1}{N} \sum_k \Omega_k^2 (G_k^m - G_k^n) \{ [n_B(\hbar\Omega_k) + 1] \exp(-i\Omega_k\tau) + n_B(\hbar\Omega_{-k}) \exp(i\Omega_{-k}\tau) \} \sum_s G_{-k}^s \langle m|s\rangle(\tau) \langle p|s\rangle^*(\tau) \quad (23)$$

$$G_p^n(t) = \int_0^{t-t_0} d\tau \frac{1}{N} \sum_k \Omega_k^2 G_k^n \{ [n_B(\hbar\Omega_k) + 1] \exp(-i\Omega_k\tau) + n_B(\hbar\Omega_{-k}) \exp(i\Omega_{-k}\tau) \} \times \sum_s G_{-k}^s \langle n|s\rangle(\tau) \langle p|s\rangle^*(\tau)$$

where

$$n_B(\hbar\Omega_k) = \frac{1}{\exp(\beta\hbar\Omega_k) - 1} \quad (24)$$

denotes the mean value of the number of phonons in the mode K being in the equilibrium state and the correlation function $\langle p|s\rangle(t)$ of the exciton subsystem is defined by

$$\langle p|s\rangle(t) = \langle p | \exp\left(-\frac{i}{\hbar} \hat{H}_e t\right) | s \rangle \quad (25)$$

with $|p\rangle$ and $|s\rangle$ denoting the exciton states localized at the sites p and s .

The time-dependent coefficients $F^m(t)$, $I_p^m(t)$, $\bar{I}_p^m(t)$ and $\tilde{I}_p^m(t)$ characterizing the influence of the exciton–photon interaction are expressed as

$$F^m(t) = \omega_{K_0} \sum_K F_K^m \langle \alpha_K(t) \rangle_f$$

$$I_p^m(t) = \int_{t_0}^t d\tau \omega_{K_0}^2 \sum_{K,K'} F_K^m \sum_s F_{-K'}^s [\langle \alpha_K(t) \alpha_{K'}^*(\tau) \rangle_f - \langle \alpha_K(t) \rangle_f \langle \alpha_{K'}^*(\tau) \rangle_f] \langle p|s\rangle^*(t-\tau)$$

$$\bar{I}_p^m(t) = \int_{t_0}^t d\tau \omega_{K_0}^2 \sum_{K,K'} F_K^m \sum_s F_{-K'}^s \{ \langle \alpha_K(t) \alpha_{K'}^*(\tau) \rangle_f - \langle \alpha_K(t) \rangle_f \langle \alpha_{K'}^*(\tau) \rangle_f + \delta_{KK'} \exp[-i\omega_K(t-\tau)] \} \langle p|s\rangle^*(t-\tau) \quad (26)$$

$$\tilde{I}_p^m(t) = \int_{t_0}^t d\tau \omega_{K_0}^2 \sum_{K,K'} F_{-K}^m \sum_s F_{-K'}^s [\langle \alpha_K^*(t) \alpha_{K'}^*(\tau) \rangle_f - \langle \alpha_K^*(t) \rangle_f \langle \alpha_{K'}^*(\tau) \rangle_f] \langle p|s\rangle^*(t-\tau).$$

The symbol $\delta_{KK'}$ denotes the Kronecker delta. New symbols for the correlation functions of the photon field operators are introduced as follows

$$\langle \alpha_K(t) \rangle_f = \text{Tr}_f \{ \hat{\rho}_f(t_0) \hat{a}_K(t-t_0) \}$$

$$\langle \alpha_K(t) \alpha_{K'}^*(\tau) \rangle_f = \text{Tr}_f \{ \hat{\rho}_f(t_0) \hat{a}_{K'}^\dagger(\tau-t_0) \hat{a}_K(t-t_0) \} \quad (27)$$

$$\langle \alpha_K^*(t) \alpha_{K'}^*(\tau) \rangle_f = \text{Tr}_f \{ \hat{\rho}_f(t_0) \hat{a}_K^\dagger(t-t_0) \hat{a}_{K'}^\dagger(\tau-t_0) \}.$$

The expressions for the time-dependent coefficients $F^m(t)$, $I_p^m(t)$, $\bar{I}_p^m(t)$ and $\tilde{I}_p^m(t)$ can be simplified and rewritten in a clearer form when we omit the influence of vacuum fluctuations of the photon field in modes which do not constitute the interaction field (for

example a pulse) and when we approximate the expressions $\epsilon_K \cdot \langle m|\hat{p}|0\rangle \exp(i\mathbf{K} \cdot \mathbf{r}_m)$ in (3) for F_K^m by $\epsilon_{K_0} \cdot \langle m|\hat{p}|0\rangle \exp(i\mathbf{K}_0 \cdot \mathbf{r}_m)$, where \mathbf{K}_0 denotes the wave vector of the most important mode in the field. This is fulfilled when all modes in the field have the same polarization vector ϵ_{K_0} and when $(\mathbf{K} - \mathbf{K}_0) \cdot \mathbf{r}_m \ll 1$ for all \mathbf{K} in the pulse and all \mathbf{r}_m in the lattice. The time-dependent coefficients $F^m(t)$, $I_p^m(t)$, $\bar{I}_p^m(t)$ and $\tilde{I}_p^m(t)$ can then be rewritten into the form

$$\begin{aligned} F^m(t) &= \omega_{K_0} \tilde{F}_{K_0}^m \mathcal{A}(t) \\ I_p^m(t) &= \int_{t_0}^t d\tau \omega_{K_0}^2 \tilde{F}_{K_0}^m \sum_s \tilde{F}_{-K_0}^s \delta N(t, \tau) \langle p|s\rangle^*(t - \tau) \\ \bar{I}_p^m(t) &= \int_{t_0}^t d\tau \omega_{K_0}^2 \tilde{F}_{K_0}^m \sum_s \tilde{F}_{-K_0}^s \delta N_v(t, \tau) \langle p|s\rangle^*(t - \tau) \\ \tilde{I}_p^m(t) &= \int_{t_0}^t d\tau \omega_{K_0}^2 \tilde{F}_{K_0}^m \sum_s \tilde{F}_{-K_0}^s \delta N_a(t, \tau) \langle p|s\rangle^*(t - \tau) \end{aligned} \quad (28)$$

with

$$\tilde{F}_{K_0}^m = -\frac{1}{\hbar \omega_{K_0}} \frac{e}{m_e} \epsilon_{K_0} \cdot \langle m|\hat{p}|0\rangle \exp(i\mathbf{K}_0 \cdot \mathbf{r}_m). \quad (29)$$

The function

$$\mathcal{A}(t) = \sum_K \sqrt{\frac{\hbar}{2\epsilon_0 V \omega_K}} \langle \alpha_K(t) \rangle_f \quad (30)$$

describes a classical amplitude of the field and noise in the field is characterized by the functions

$$\begin{aligned} \delta N(t, \tau) &= \sum_{K, K'} \frac{\hbar}{2\epsilon_0 V \sqrt{\omega_K \omega_{K'}}} \langle \delta \alpha_K(t) \delta \alpha_{K'}^*(\tau) \rangle_f \\ \delta N_v(t, \tau) &= \delta N(t, \tau) + \sum_K \frac{\hbar}{2\epsilon_0 V \omega_K} \exp[-i\omega_K(t - \tau)] \\ \delta N_a(t, \tau) &= \sum_{K, K'} \frac{\hbar}{2\epsilon_0 V \sqrt{\omega_K \omega_{K'}}} \langle \delta \alpha_K^*(t) \delta \alpha_{K'}^*(\tau) \rangle_f \end{aligned} \quad (31)$$

where $\delta \alpha_K(t) = \alpha_K(t) - \langle \alpha_K(t) \rangle_f$. The description of the photon field statistics is restricted to the first and second moments which is a consequence of the perturbation approximation in the exciton–photon coupling constants which was invoked.

Equations (20)–(22) for the matrix elements of the exciton-reduced density matrix $\hat{\rho}_e(t)$ represent a generalization of the standard approaches in the direction of incorporation of effects connected with processes of excitation creation and annihilation under the influence of an ultrashort pulse. Their comparison with the standard approaches is done in the next section. Now we would like to bring to the reader's attention some interesting properties of the above equations.

The time-dependent coefficients in the resulting equations are exact to the second order in the exciton–photon and the exciton–phonon coupling. Owing to this restriction and owing to the chaotic statistics of phonons there are no terms which would combine the influence of both the exciton–photon and the exciton–phonon interaction on the exciton subsystem. Inclusion of such terms is important for strong photon fields [3].

If we restrict our considerations to classical deterministic fields, then the time-dependent coefficients $I_p^m(t)$, $\bar{I}_p^m(t)$ and $\tilde{I}_p^m(t)$ proportional to the second power of the exciton–photon

coupling constants are zero. We suppose that terms proportional to the third and higher powers of the exciton–photon coupling constants are zero in this case, too. This assumption can be verified for a dimer [11].

The matrix elements of the exciton reduced density matrix $\rho_{mm}^e(t)$ and $\rho_{00}^e(t)$ fulfil the relation

$$\sum_m \rho_{mm}^e(t) + \rho_{00}^e(t) = 1 \quad (32)$$

following from equations (20) and (21) which expresses the conservation law of the ‘total probability’.

Although equations (20)–(22) are primarily established for the description of the excitation dynamics in large exciton systems with the interaction with phonons under the influence of an ultrashort pulse, they can also be conveniently used for the description of simple systems like dimers. In fact, equations (20)–(22) in the case of a dimer represent a strong generalization of the Bloch equations [11, 28]. This generalization is in three respects: equations for the dimer incorporate effects of the transfer of excitation to the dynamics of an atom (molecule) exposed to a photon field, they describe the interaction with a phonon reservoir more precisely and more flexibly and they are able to describe the influence of quantum noise in the photon field. A detailed study of the time development of an excitation in the dimer reveals very interesting effects, especially in the region of values of parameters when times characterizing pulse duration, propagation of the excitation and its relaxation are comparable [11, 28].

The above derived equations can be used for investigations of more complex exciton systems, e.g. polymeric chains.

5. Comparison with standard theories

The standard theories based either on time-convolution generalized master equations or on time-convolutionless ones do not take into account the interaction with an ultrashort optical pulse. Instead, they suppose an initially created excitation at a given place in the lattice. In order to obtain a comparison of our approach with the standard theories, we have to investigate our equations for cases when the optical field does not already affect the exciton system (vacuum fluctuations of the photon field, i.e. spontaneous exciton decay processes, are also supposed to be negligible henceforth).

The set of coupled equations (20)–(22) for $\rho_{mn}^e(t)$, $\rho_{0n}^e(t)$ and $\rho_{00}^e(t)$ decomposes into two sets of coupled equations for $\rho_{mn}^e(t)$, $\rho_{00}^e(t)$ and for $\rho_{0n}^e(t)$ in this case,

$$\frac{d}{dt} \rho_{mn}^e(t) = -\frac{i}{\hbar} \sum_p J_{mp} \rho_{pn}^e(t) + \frac{i}{\hbar} \sum_p J_{pn} \rho_{mp}^e(t) - \sum_p G_p^{mn}(t) \rho_{pn}^e(t) - \sum_p G_p^{nm*}(t) \rho_{mp}^e(t) \quad (33)$$

$$\frac{d}{dt} \rho_{00}^e(t) = 0 \quad (34)$$

$$\frac{d}{dt} \rho_{0n}^e(t) = \frac{i}{\hbar} \sum_p J_{pn} \rho_{0p}^e(t) - \sum_p G_p^{n*}(t) \rho_{0p}^e(t). \quad (35)$$

It follows from (34) that $\rho_{00}^e(t)$ remains constant and relation (32) implies that $\sum_m \rho_{mm}^e(t)$ is constant and less than or equal to 1 (in contrast to [12–14] where one exciton is assumed to be present in the system for all times, i.e. $\sum_m \rho_{mm}^e(t) = 1$).

The set of equations (33) for the matrix elements $\rho_{mn}^e(t)$ is precisely that one obtained from the time-convolutionless generalized master equations applied to the exciton–phonon

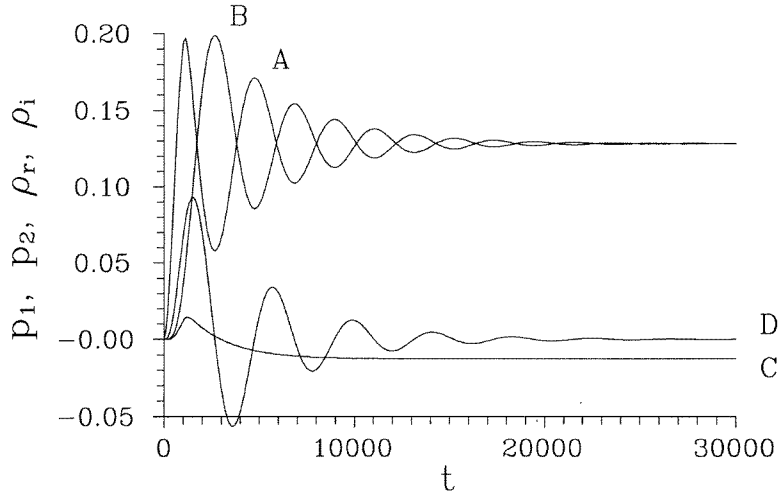


Figure 1. The probabilities [A] $p_1(t)$ ($p_1 = \rho_{11}^e$) and [B] $p_2(t)$ ($p_2 = \rho_{22}^e$) and the elements [C] $\rho_r(t)$ and [D] $\rho_i(t)$ ($\rho_{12}^e = \rho_r + i\rho_i$) for the dimer; $F_1 = 0.0005$, $F_2 = 0$, $J = 0.0005$, $\varepsilon = 0$, $\hbar\delta' = 0$, $\tau_1 = 1000$, $\tau_2 = 200$, $G = 0.005$, $G_1/G = 1 + 0.25i$, $G_2/G = 1 - 0.25i$, $\hbar\Omega_{\text{ph}} = 0.01$, $\hbar\gamma_{\text{ph}} = 0.01$, $n_B = 0$; for details see [11, 28] (quantities of energy are in eV, time is in fs).

system [12–14]. However, there is a question: What is the exciton-reduced density matrix at the end of the interaction with an optical pulse? Our theory shows that, in general, a pulse leaves the exciton subsystem in a state which cannot be described by a density matrix with zero off-diagonal matrix elements. This is clearly demonstrated in the case of a dimer in figure 1 [11]. Nonzero off-diagonal matrix elements at the time when the pulse switches off, mean that the probabilities (diagonal matrix elements) do not evolve in time according to $(t - t_f)^2$ (t_f being the time at which the pulse sharply switches off) in the short period after the pulse switches off (i.e. there is no ‘slippage time’). This also means, that the excitation is more delocalized after the pulse switches off and thus has a lower kinetic energy. This leads to a faster loss of the exciton coherence.

Thus the standard theories working only with diagonal matrix elements of the exciton-reduced density matrix, have a very limited area of application, because they also suppose initially zero off-diagonal matrix elements. The assumption of zero off-diagonal matrix elements is inevitable in this case because the initial condition term must be precisely zero in order to keep consistency of the perturbation approximation invoked in the derivation of equations. In principle the standard theories working with off-diagonal matrix elements [12–14] are also able to describe the excitation dynamics well, but there is a question of how to determine the initial exciton-reduced density matrix. The above developed theory does not need it.

The time-dependent coefficients $G_p^{mn}(t)$ and $G_p^n(t)$ in (23) can be further simplified for times much longer than the relaxation time τ_R of the phonon reservoir, which is important

for the long-time analysis of the excitation behaviour,

$$\begin{aligned}
 G_p^{mn} &= \lim_{t \rightarrow \infty} G_p^{mn}(t) = -\frac{i\hbar}{N} \sum_k \Omega_k^2 (G_k^m - G_k^n) \sum_s G_{-k}^s \sum_{\nu_1, \nu_2} \left[\frac{n_B(\hbar\Omega_k) + 1}{E_{\nu_1} - E_{\nu_2} + \hbar\Omega_k - i\epsilon} \right. \\
 &\quad \left. + \frac{n_B(\hbar\Omega_{-k})}{E_{\nu_1} - E_{\nu_2} - \hbar\Omega_{-k} - i\epsilon} \right] \langle m | \nu_1 \rangle \langle \nu_1 | s \rangle \langle s | \nu_2 \rangle \langle \nu_2 | p \rangle \\
 G_p^n &= \lim_{t \rightarrow \infty} G_p^n(t) = -\frac{i\hbar}{N} \sum_k \Omega_k^2 G_k^n \sum_s G_{-k}^s \sum_{\nu_1, \nu_2} \left[\frac{n_B(\hbar\Omega_k) + 1}{E_{\nu_1} - E_{\nu_2} + \hbar\Omega_k - i\epsilon} \right. \\
 &\quad \left. + \frac{n_B(\hbar\Omega_{-k})}{E_{\nu_1} - E_{\nu_2} - \hbar\Omega_{-k} - i\epsilon} \right] \langle n | \nu_1 \rangle \langle \nu_1 | s \rangle \langle s | \nu_2 \rangle \langle \nu_2 | p \rangle
 \end{aligned} \tag{36}$$

where E_ν denotes the eigenenergy corresponding to the eigenstate $|\nu\rangle$ of the free exciton Hamiltonian \hat{H}_e ; ϵ denotes a small parameter which goes to zero after performing all calculations (integration over k after substituting sums over k by integrals in (36)). The coefficients G_p^{mn} in (36) were already derived in [29] when applying the time-convolutionless generalized master equations to exciton–phonon systems. It was shown for simple systems, that they describe correctly the time evolution of an excitation towards the equilibrium state. Such coefficients can also be derived when the Born–Markov approximation in the interaction picture in the time-convolution generalized master equations is invoked. The use of time-convolution equations leads to unphysical results in other cases [22].

6. Conclusions

Starting from the Liouville equation for the density matrix we have developed a microscopic theory which allows us to treat simultaneous creation, annihilation, propagation and relaxation of an excitation in molecular condensates. It generalizes standard approaches which are based on the assumption, that the excitation is initially created at a given place in the system. We have shown that the standard approaches, not including off-diagonal matrix elements of the exciton-reduced density matrix, have a very limited area of application, because off-diagonal elements are generally nonzero once the pulse switches off. This means that the time delay known as a ‘slippage time’ does not occur. Also the loss of exciton coherence is faster.

The above developed general theory opens the possibility to study exciton systems with such values of parameters, that the mutual interplay of creation, annihilation, propagation and relaxation of an excitation plays an important role.

The theory is also suitable for investigations of the influence of statistical properties of optical fields (including nonclassical ones) interacting with exciton systems.

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