Hybrid density matrix approach as a factorization scheme for many-body systems: Illustrated by a quantum dot–continuum system

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We present a theoretical factorization scheme for interactions between many-body systems with localized discrete and delocalized quasicontinuous states. The projection operator technique combines a conventional correlation expansion with an exact diagonalization scheme. Our approach is capable of treating the continuum dynamically and to apply non-Markovian treatments beyond the Markovian approximation typically used in Lindblad formalism. Therefore it can be an important tool to describe a variety of systems as well as interaction mechanisms.

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I. INTRODUCTION

A variety of many-body systems exists that consists of a subsystem with localized, discrete states and a subsystem with quasicontinuous states. One important example is given by semiconductor quantum dots (QDs) coupled to the embedding bulk, quantum well, or wetting layer carrier reservoir. Beside self-organized QDs, also other systems such as impurities in bulk or other lower dimensional structures like quantum wells [1–3] and wires [4], or molecule systems [5–9] form such a hybrid system since a finite number of discrete, localized states is interacting with a continuum of delocalized states.

Both subsystems are often coupled via Coulomb interaction, but also by other coupling mechanisms like electron-phonon interaction [10–12]. These interactions include scattering processes between the two subsystems. The factorization scheme presented in this paper is constructed to describe the coupling and scattering dynamics between these hybrid many-body systems. To illustrate the principle of the theoretical scheme, we choose in this paper the example of QDs embedded in a carrier reservoir, due to their fundamental role in various applications, such as single photon emitters [13,14], conventional laser diodes, microcavity lasers [15,16] or efficient nonclassical light sources [17].

In semiconductor QD devices, the in-scattering of carriers into the QD-like electrons and holes typically occurs from the delocalized continuum states of a carrier reservoir. Therefore carrier scattering processes into discrete QD states are relevant in a variety of applications such as intermediate band solar cells [18], infrared emitters [19], and photodetectors [3,20] and have been investigated extensively by experiments [21–24], for instance, via two-color pump-probe spectroscopy [21,22] as well as in theoretical studies [25–32].

Both systems (in our example, carrier reservoir and localized, discrete QD states) constitute many-body systems of different type and must be described by different approximations. The reservoirs exhibit a high density of (occupied) continuous states, especially in the case of electrical pumping. Here, fluctuations in comparison to the average occupation number are of minor importance and, typically, conventional cluster correlation expansions are used and provide an accurate description of the physical effects [31,33].

In contrast, localized few-particle configurations in the QD, such as excitons, trions, and biexcitons exhibit strong fluctuations with respect to mean occupation numbers. Therefore it is necessary to describe the discrete states inside the QDs without using correlation or cluster expansion, but with configurations like the ground state, single, bi-, or more exciton and trion states, etc., describing the different configurations [27–29].

We aim to treat both systems by different approximations, but like to also include the coupling for describing the (correlated) electron transfer from the reservoir to the QD. For such a configuration, the continuum is typically treated as a bath in the Lindblad approach [27–31]. Especially, Jahnke *et al.* published some interesting results of carrier scattering processes in QDs beyond the Boltzmann equation using Lindblad formalism [28]. However, the treatment, so far, is restricted to the Markovian treatment using Lindblad formalism and to a description of the carrier reservoir as a bath.

In this paper, we present a theoretical scheme, which combines the advantages of both approaches (correlation expansions and excitonic states) into a single consistent expansion scheme: therefore the QD configurations are solved by an exact diagonalization scheme, similar to the method used in Ref. [27–29,31]. This way, correlations (e.g., Coulomb induced) in the localized QD-states are treated nonperturbatively. However, extending the treatment of these references, we treat the correlations of the continuum reservoir states without using bath approximation in Lindblad formalism, but in a mean field approach using single particle occupations as first order for a cluster based description of the continuum correlations [34]. This way the approach is capable of treating the continuum dynamically and to go beyond the Markovian approximation. Especially for a population of the reservoir via external electrical contacts, the presented hybrid density matrix approach provides a method to determine its dynamics in response to modulations of the applied current (important, e.g., for application in single photon regime). The resulting equations are derived using a projection operator technique and can be solved in different orders of approximation for the reservoir-QD coupling. To compare our results with literature, we consider the limit of a Markovian treatment of the Coulomb coupling in second-order Born approximation for the QD-reservoir scattering. The equations give similar results for the limit of treating the continuum as bath as the Lindblad approach of Refs. [28,29,32].

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The method is applicable for various systems and for different interaction processes such as Coulomb coupling or electron-phonon interaction, e.g., inducing spin-flip processes. For simplicity, the example focusses only on the Coulomb interaction.

II. PROJECTION OPERATOR FORMALISM

As a model system we use a QD with N_e discrete spin-degenerated states x in the conduction and N_h states y in the valence band embedded in quasicontinuous states ν, e.g., a wetting layer or bulk material. A fermionic many-particle Fock state of the QD with the occupation number $n^{e(h)}_{x(y)} = 0,1$ for electrons (holes) can be constructed using

$$|\mathbf{n}_{I_eI_h}\rangle = \left|n^e_1,\ldots,n^e_{N_e},n^h_1,\ldots,n^h_{N_h}\right\rangle = e^\dagger_{x_1}\ldots e^\dagger_{x_{I_e}} h^\dagger_{y_1}\ldots h^\dagger_{y_{I_h}}|\phi_0\rangle, \quad (1)$$

from the ground state $|\phi_0\rangle$ of the QD system with no electron or hole carrier in the conduction or valence band, respectively. Here, $e^\dagger_x$ (e_x) denote the creation (annihilation) operators for electrons of the QD state x and $h^\dagger_y$ (h_y) for holes of the QD state y. The QD is populated with $n^e_1+\ldots+n^e_{N_e} = I_e$ electrons and $n^h_1+\ldots+n^h_{N_h} = I_h$ holes in state $|\mathbf{n}_{I_eI_h}\rangle$. For the wetting layer, the description via Fock states works in an analog way. $|\mathbf{n}_{I_eI_h},\boldsymbol{\nu}\rangle$ denotes a complete system state, consisting of a set of QD states $\mathbf{n}_{I_eI_h}$ with I_e electrons and I_h holes and wetting layer states $\boldsymbol{\nu}$. The projection operator $P^{I_eI_h}$ projecting into these many particle states with I_e electrons and I_h holes is given by

$$P^{I_eI_h} = \sum_{\mathbf{n}_{I_eI_h},\boldsymbol{\nu}} |\mathbf{n}_{I_eI_h},\boldsymbol{\nu}\rangle\langle\mathbf{n}_{I_eI_h},\boldsymbol{\nu}|. \quad (2)$$

In the projection operator formalism the description of the QD configurations can be formulated via a local QD operator $L^{I_eI_h\dagger}_{\mathbf{i}}$:

$$L^{I_eI_h\dagger}_{\mathbf{i}} \equiv P^{I_eI_h} e^\dagger_{x_1}\ldots e^\dagger_{x_{I_e}} h^\dagger_{y_1}\ldots h^\dagger_{y_{I_h}} P^{00}, \quad (3)$$

with the multi-index $\mathbf{i} := \{x_1\ldots x_{I_e}, y_1\ldots y_{I_h}\}$ (note that $P^{00} = |\phi_0\rangle\langle\phi_0|$). The operators $L^{I_eI_h\dagger}_{\mathbf{i}}$ are the analog to bra-ket states in the isolated QD systems. Due to these local QD creation (annihilation) operators $L^{I_eI_h\dagger}_{\mathbf{i}}$ ($L^{I_eI_h}_{\mathbf{i}}$), all localized few particle configurations in the QD, such as excitons, trions, and biexcitons can be represented. In Fig. 1(b), several QD configurations are depicted as well as the interaction between the QD and the carrier reservoir, cf. Fig. 1(a).

III. HAMILTONIAN

The dynamics of the interacting QD-continuum system are calculated by Heisenberg equations of motion. For our example, the total Hamiltonian H consists of the single-particle contribution H_0 and the Coulomb-interaction H_C:

$$H_0 = \sum_{n,\mu}\epsilon^{\lambda_\mu}_n \mu^\dagger_n\mu_n + \sum_{\nu,\mu}\epsilon^{\lambda_\mu}_\nu \mu^\dagger_\nu\mu_\nu, \quad (4)$$

where $\epsilon^{\lambda_\mu}_{n(\nu)}$ is the single-particle energy of the bound QD states n and of the unbound continuum states ν with the band index $\lambda_\mu \in \{c,v\}$. For compact notation, we introduce a carrier

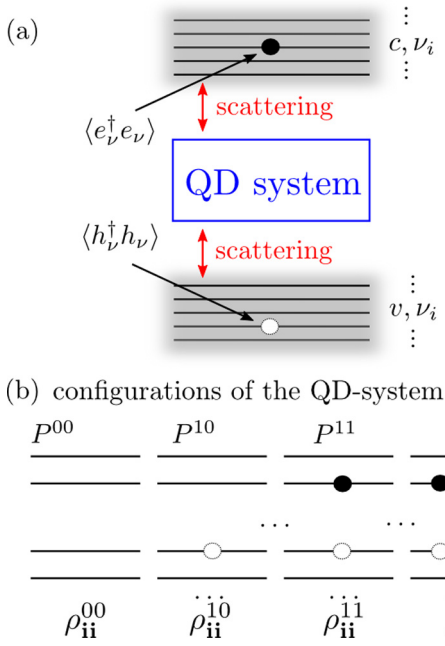

FIG. 1. (Color online) (a) Level scheme of the QD-continuum system. (b) Several configurations of the electron and hole occupations in the QD system. Here, the diagonal elements of the QD density matrix $\rho^{I_eI_h}_{\mathbf{ii}}$ are presented. The developed theoretical scheme is also applicable to nondiagonal contributions.

index $\mu \in \{e,h\}$. The Coulomb interaction using the envelope approximation for the single-particle wave functions is given in electron hole picture by

$$H_C = \frac{1}{2}\sum_{\mathbf{s},\mu_1,\mu_2} V^{s_1s_2s_3s_4}_{\mu_1\mu_2}\mu_{1s_1}^\dagger\mu_{2s_2}^\dagger\mu_{2s_3}\mu_{1s_4}, \quad (5)$$

where s_1,s_2,s_3,s_4 are multi-indices of the energy levels of the system, consisting of index n for bound QD states or continuum states ν and spin σ. Here, we consider the part of Coulomb interaction between two types of carriers μ_1 and μ_2. The Coulomb coupling elements $V^{s_1s_2s_3s_4}_{\mu_1\mu_2}$ of the Coulomb potential $W(\mathbf{r},\mathbf{r}')$ read

$$V^{s_1s_2s_3s_4}_{\mu_1\mu_2} = q_{\mu_1}q_{\mu_2}\int\int \phi^*_{\mu_1,s_1}(\mathbf{r})\phi^*_{\mu_2,s_2}(\mathbf{r}')W(\mathbf{r},\mathbf{r}') \times \phi_{\mu_2,s_3}(\mathbf{r}')\phi_{\mu_1,s_4}(\mathbf{r})d\mathbf{r}d\mathbf{r}'\delta_{\sigma_{s_1},\sigma_{s_4}}\delta_{\sigma_{s_2},\sigma_{s_3}}, \quad (6)$$

where $\phi_{\mu,s}(\mathbf{r})$ denote the wave functions and σ the spin.

Especially, for high carrier densities of the continuum, the capture dynamics within the QD-continuum system can be assumed to be dominated by Coulomb scattering [35]. Here, we neglect the influence of electron-phonon processes for the example used to demonstrate the scheme, but the factorization scheme is also applicable for electron-phonon scattering.

The QD in- and out-scattering result from Auger type processes between the localized QD- and continuous reservoir states. There are two different types of Coulomb induced scattering processes both illustrated in Fig. 2: the capture of one carrier into a QD state and carrier relaxation within the reservoir (reservoir assisted) or within the QD (QD assisted).

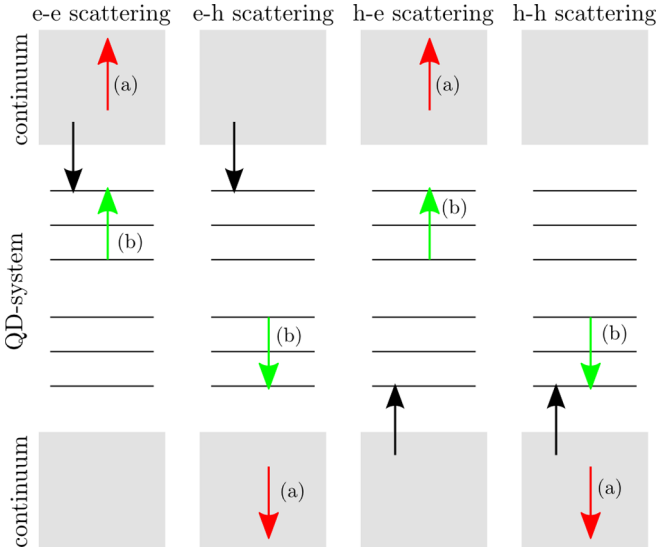

FIG. 2. (Color online) Coulomb induced electron and hole capture via Auger processes between bound QD and continuous states of the carrier reservoir: (a) carrier relaxation within the reservoir or (b) within the QD.

This way, the energy conservation for the entire process is balanced [32,35,36].

IV. EXCITON ENERGY EIGENSTATES

Due to the discrete nature of QD states, Coulomb correlations inside QDs can be very strong. In contrast to continua with a high number of states and high electron occupations, QDs have only a few discrete states, which are energetically well separated. Consequently, the QD is populated by a small number of carriers. Varying configurations of the carriers inside the QD lead to significant differences of the Coulomb interaction and distinct energies of the discrete configurations. Therefore it is useful to describe the discrete QD states by excitonic states, which can be diagonalized with respect to the internal Coulomb interaction H^{QD}:

$$\left(H_0^{\mathrm{QD}} + H_C^{\mathrm{QD}}\right) X_{\mathbf{j}}^{I_e I_h \dagger} = E_{\mathbf{j}}^{I_e I_h} X_{\mathbf{j}}^{I_e I_h \dagger}, \tag{7}$$

where we transform the local states inside the QD into new eigenstates:

$$X_{\mathbf{j}}^{I_e I_h \dagger} = \sum_{\mathbf{j}} c_{\mathbf{ij}}^{I_e I_h *} L_{\mathbf{i}}^{I_e I_h \dagger}, \tag{8}$$

so that they are eigenstates of the QD contribution with the eigenenergies $E_{\mathbf{j}}^{I_e I_h}$ and the expansion coefficients $c_{\mathbf{ij}}^{I_e I_h *}$. Similar to the local QD operators, the Coulomb coupling elements are redefined into effective coupling elements $\tilde{V}$ with respect to the expansion coefficients for continuum assisted scattering:

$$\tilde{V}^{\nu_1 \nu_2 \nu_3}_{\substack{\mathbf{ij},\mu_1\mu_2 \\ I_e I_h}} = \sum_{\mathbf{a}} \sum_{n_1} c_{\mathbf{a,j}}^{I_e I_h *} V^{n_1 \nu_1 \nu_2 \nu_3}_{\mu_1 \mu_2} c_{\mathbf{i,a}n_1}^{I_e+1 I_h}, \tag{9}$$

and QD assisted scattering:

$$\tilde{V}^{\nu_1}_{\substack{\mathbf{ij},\mu_1\mu_2 \\ I_e I_h}} = \sum_{\mathbf{a}} \sum_{n_1,n_2,n_3} c_{\mathbf{a,j}}^{I_e I_h *} V^{n_1 n_2 n_3 \nu_1}_{\mu_1 \mu_2} c_{\mathbf{i,a}n_1 n_2/n_3}^{I_e+1 I_h}. \tag{10}$$

V. HYBRID DENSITY MATRIX EQUATION

A. Observables

The observables of central interest to characterize the system are given by the electron and hole occupations $f_\nu^\mu = \langle \mu_\nu^\dagger \mu_\nu \rangle$ in the first order of the continuum state ν (polarizations $p^\mu_{\nu_1,\nu_2} = \langle \mu_{\nu_1}^\dagger \mu_{\nu_2} \rangle$) as well as by the reduced density matrix of the QD system $\rho_{\mathbf{ij}}^{I_e I_h, I'_e I'_h}$ (**i**,**j** denote the QD configurations, I_e the electron and I_h the hole number):

$$\rho_{\mathbf{ij}}^{I_e I_h, I'_e I'_h} = \left\langle P^{I_e I_h} X_{\mathbf{i}}^{I_e I_h \dagger} X_{\mathbf{j}}^{I'_e I'_h} P^{I'_e I'_h} \right\rangle, \tag{11}$$

where $P^{I_e I_h}$ is a projection operator and $X_{\mathbf{i}}^{I_e I_h \dagger}$ is the creation operator of the QD energy eigenstates. Note that for the QD states, the diagonalized many-body configurations enter, which is of particular importance for high (biexciton) binding energies found in nitride QDs [37,38].

The temporal evolution of the reduced QD density matrix $\rho_{\mathbf{ij}}^{I_e I_h, I'_e I'_h}$ and the continuum occupations f_ν^μ with respect to Auger processes are calculated via the Heisenberg equation of motion approach [39]. For illustrative purposes, the resulting hierarchy of equations of motion is illustrated in Fig. 3 for QD assisted electron-electron scattering. The dynamics of the

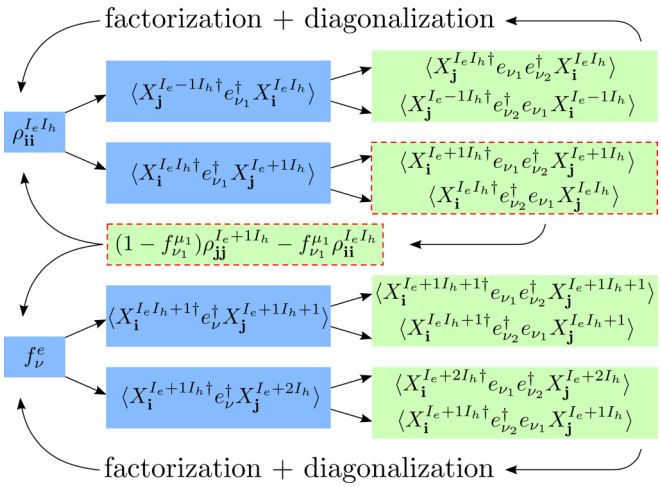

FIG. 3. (Color online) Scheme of the hybrid density matrix approach: the temporal evolution of the system observables describing the occupation probability of the QD $\rho_{\mathbf{i}}^{I_e I_h}$ and the continuum f_ν^e is determined by assisted quantities of QD and continuum contributions. To solve the hierarchy problem, the assisted expectation values can be factorized in the second order of equation of motion. Here, we present diagonal elements of the QD density matrix and of the reservoir. However, the hybrid density matrix approach can be also applied to nondiagonal elements. For illustrative purposes, only electron-electron scattering processes including carrier relaxation within the QD, shown in Fig. 2, left (b), are depicted. For all other electron and hole capture processes, in particular, containing reservoir relaxation, the principle of the scheme is the same. (The hole continuum can be described identical to the electron continuum.)

QD density matrix $\rho_{\mathbf{ij}}^{I_e I_h, I'_e I'_h}$ [cf. Eq. (A1)] and the continuum densities f_ν^μ [cf. Eq. (A2)] (polarizations p_{ν_1,ν_2}^μ) couples to assisted expectation values consisting of QD and continuum contributions such as $\langle X_{\mathbf{i}}^{I_e I_h \dagger} e_{\nu_1}^\dagger X_{\mathbf{j}}^{I_e+1 I_h} \rangle$ [cf. blue box in Fig. 3 and Eq. (A3)] whose temporal evolution themselves depends on assisted quantities of higher order, cf. Appendix and green box in Fig. 3. Quantities of always increasing number of continuum fermionic operators occur during the derivation of the hierarchy and the set of equation of motion does not close, which is typically called hierarchy problem [40]. Factorization rules provide a method to close the set of equation of motions.

B. Factorization Scheme

To derive a suitable factorization scheme, we assume that for the continuum a description using single particle observables is sufficient. (Note that similar schemes can be derived for closing the scheme at two particle correlations like $\langle e_{\nu_1}^\dagger e_{\nu_2}^\dagger e_{\nu_3} e_{\nu_4} \rangle^c$ [cf. Eq. (14)] in the continuum.) In contrast, we have to consider all occupied states inside the QD. This is equivalent to a treatment of the continuum in Hartree-Fock level and for the QD beyond correlation expansion level.

These two assumptions lead to the following assumed form of the statistical operator $\rho(t) = \rho_{\mathrm{QD}}(t)\rho_{\mathrm{ct.}}(t)$ including QD and continuum (ct.) contributions:

$$\begin{aligned}\rho_{\mathrm{ct.}}(t) =& \frac{1}{Z_{\mathrm{ct.}}} \exp\Bigg[- \sum_{\nu,\eta} \left(\alpha_{\nu,\eta}^{ee}(t) e_\nu^\dagger e_\eta + \alpha_{\nu+K,\eta+K}^{hh}(t) h_{\nu+K} h_{\eta+K}^\dagger \right) \\ &+ \alpha_{\nu+K,\eta}^{eh}(t) e_{\nu+K}^\dagger h_\eta^\dagger + \alpha_{\nu,\eta+K}^{he}(t) h_\nu e_{\eta+K} \Bigg], \end{aligned} \tag{12}$$

$$\rho_{\mathrm{QD}}(t) = \frac{1}{Z_{\mathrm{QD}}} \exp\left(- \sum_{\mathbf{i},\mathbf{j}, I_e, I_h, I'_e, I'_h} \beta_{\mathbf{i},\mathbf{j}}^{I_e I_h, I'_e I'_h}(t) X_{\mathbf{i}}^{I_e I_h \dagger} X_{\mathbf{j}}^{I'_e I'_h} \right), \tag{13}$$

as basis for the factorization rules [41] with the partition functions Z_{QD} and $Z_{\mathrm{ct.}}$, the number of continuum states K and the Lagrange parameters $\alpha_{\nu,\eta}^{\mu_1,\mu_2}(t)$ and $\beta_{\mathbf{i},\mathbf{j}}^{I_e I_h, I'_e I'_h}(t)$. It can be shown, that the continuum expectation values factorize identical to the Hartree-Fock-factorization. For two-particle continuum quantities, the factorization rule reads

$$\begin{aligned}\langle \mu_{\nu_1}^\dagger \mu_{\nu_2}^\dagger \mu_{\nu_3} \mu_{\nu_4} \rangle =& \langle \mu_{\nu_1}^\dagger \mu_{\nu_4} \rangle \langle \mu_{\nu_2}^\dagger \mu_{\nu_3} \rangle - \langle \mu_{\nu_1}^\dagger \mu_{\nu_3} \rangle \langle \mu_{\nu_2}^\dagger \mu_{\nu_4} \rangle \\ &+ \langle \mu_{\nu_1}^\dagger \mu_{\nu_2}^\dagger \mu_{\nu_3} \mu_{\nu_4} \rangle^c, \end{aligned} \tag{14}$$

where $\langle \mu_{\nu_1}^\dagger \mu_{\nu_2}^\dagger \mu_{\nu_3} \mu_{\nu_4} \rangle^c$ denotes correlations. The resulting factorization rule for assisted expectation values, consisting of continuum contributions $A^\dagger A$ and QD contributions $X^\dagger X$, read

$$\langle A^\dagger A X^\dagger X \rangle = \langle A^\dagger A \rangle \langle X^\dagger X \rangle + \langle A^\dagger A X^\dagger X \rangle^c. \tag{15}$$

As a first approximation, we neglect the correlations $\langle \mu_{\nu_1}^\dagger \mu_{\nu_2}^\dagger \mu_{\nu_3} \mu_{\nu_4} \rangle^c$ and $\langle A^\dagger A X^\dagger X \rangle^c$ (second-order Born level).[1]

However, in analogy to conventional correlation expansion [40], our theoretical approach can be expanded to these correlations as well.

The factorization scheme is in principle a treatment in the second-order Born approximation [42]. The factorization is the key element to treat the reservoir as a dynamical variable and not as a bath in thermodynamic equilibrium via Lindblad terms.[2] The factorization rule is restricted to expectation values featuring the same number of creation and annihilation operators of the continuum. It is an open question whether other statistical operators can be found resulting in factorization rules, which can be already used for lower order assisted expectation values such as $\langle X^\dagger A^\dagger X \rangle$ (where $A^\dagger$ is a continuum operator) similar to the Fock term.

First, we apply the factorization rule (15) to the mixed expectation values like $\langle X_{\mathbf{i}}^{I_e I_h \dagger} e_{\nu_1}^\dagger e_{\nu_2}^\dagger e_{\nu_3} e_{\nu_4} X_{\mathbf{j}}^{I'_e I'_h \dagger} \rangle$ (cf. Fig. 3 green box) in the equations of motion (A3)–(A5) to separate QD $\langle X_{\mathbf{i}}^{I_e I_h \dagger} X_{\mathbf{j}}^{I'_e I'_h \dagger} \rangle$ and continuum contributions $\langle e_{\nu_1}^\dagger e_{\nu_2}^\dagger e_{\nu_3} e_{\nu_4} \rangle$. Subsequently, we factorize the continuum quantities such as $\langle e_{\nu_1}^\dagger e_{\nu_2}^\dagger e_{\nu_3} e_{\nu_4} \rangle$ via Hartree-Fock, e.g., using Eq. (14).

For simplicity and because they are often discussed in literature in master equations, we present in this paper only the diagonal elements of the QD density matrix $\rho_{\mathbf{ii}}^{I_e I_h, I'_e I'_h} \equiv \rho_{\mathbf{i}}^{I_e I_h}$ and of the reservoir f_ν^μ, neglecting fast decaying coherences such as $\langle e_{\nu_1}^\dagger h_{\nu_2}^\dagger \rangle$, p_{ν_1,ν_2}^μ and $\rho_{\mathbf{ij}}^{I_e I_h, I'_e I'_h}$. However, the developed theoretical scheme provides equations of nondiagonal contributions like coherences as well. Structurally, these equations are very similar to the equations presented in this paper.

The factorization rules result in a closed set of equations of motion, consisting of assisted quantities such as $\langle X_{\mathbf{i}}^\dagger e_\nu^\dagger X_{\mathbf{j}} \rangle$ and system observables like $\rho_{\mathbf{i}}^{I_e I_h}$. The derived hybrid density matrix equations resemble a mixture of density matrix contributions for the occupation probability of QD configurations $\rho_{\mathbf{i}}^{I_e I_h}$ and typical Boltzmann equation like features for the average populations in the reservoir f_ν^μ, including their Pauli blocking. The complete non-Markovian set of equations is presented in the Appendix, Eqs. (A1)–(A5).

C. Markovian Limit

For a first insight, we solve the equations of the assisted quantities Eqs. (A3)–(A5) via a Markovian treatment. The important part for describing the dynamics of the emission process is given by the reduced density matrix of the QD:

$$\begin{aligned}\partial_t \rho_{\mathbf{i}}^{I_e I_h} =& \frac{2\pi}{\hbar^2} \sum_{\mathbf{j},\nu_1} \Bigg[T_{\substack{\mathbf{ij},ee \\ I_e+1;I_h}}^{\nu_1} - T_{\substack{\mathbf{ji},ee \\ I_e;I_h}}^{\nu_1} + T_{\substack{\mathbf{ij},eh \\ I_e+1;I_h}}^{\nu_1} \\ &- T_{\substack{\mathbf{ji},eh \\ I_e;I_h}}^{\nu_1} + \sum_{\nu_2,\nu_3} \Bigg(S_{\substack{\mathbf{ij},ee \\ I_e+1;I_h}}^{\nu_1\nu_2\nu_3} - S_{\substack{\mathbf{ji},ee \\ I_e;I_h}}^{\nu_1\nu_2\nu_3} \\ &+ S_{\substack{\mathbf{ij},eh \\ I_e+1;I_h}}^{\nu_1\nu_2\nu_3} - S_{\substack{\mathbf{ji},eh \\ I_e;I_h}}^{\nu_1\nu_2\nu_3} \Bigg) \Bigg] + e \leftrightarrow h, \end{aligned} \tag{16}$$

[1] Note that due to the exact factorization in the QD system we are beyond Hartree-Fock level.

[2] In the description of electron-phonon interaction, similar approaches are typically used: the influence of phonons can be approximated via Lindblad terms or dynamically, e.g., in the second-order Born approximation.

where $e \leftrightarrow h$ means that all listed contributions enter again with exchanged carrier indices e and h. The full strength of the hybrid approach is the accessibility of the average populations in the reservoir f_ν^μ as a new dynamical variable[3]:

$$\partial_t f_\nu^e = \frac{2\pi}{\hbar^2} \sum_{I_e, I_h, \mathbf{i},\mathbf{j}} \Bigg[T^{\nu}_{\substack{\mathbf{ij},ee \\ I_e+2;\, I_h}} + T^{\nu}_{\substack{\mathbf{ij},eh \\ I_e+1;\, I_h+1}} + \sum_{\nu_1,\nu_2} \Bigg(S^{\nu\nu_1\nu_2}_{\substack{\mathbf{ij},ee \\ I_e+1;\, I_h}} - S^{\nu_2\nu_1\nu}_{\substack{\mathbf{ij},ee \\ I_e+1;\, I_h}} + S^{\nu\nu_1\nu_2}_{\substack{\mathbf{ij},eh \\ I_e+1;\, I_h}} - S^{\nu_2\nu_1\nu}_{\substack{\mathbf{ij},he \\ I_e;\, I_h+1}} + S^{\nu_1\nu\nu_2}_{\substack{\mathbf{ij},he \\ I_e;\, I_h+1}} \Bigg) \Bigg]. \tag{17}$$

The hole occupation has the same form just the carrier index e is changed to h. According to Fig. 2, the Coulomb scattering can be distinguished in carrier capture processes including carrier relaxation within the reservoir [36]:

$$S^{\nu_1\nu_2\nu_3}_{\substack{\mathbf{ij},\mu_1\mu_2 \\ I_e+1;\, I_h(+1)}} = \left| \tilde{V}^{\nu_3\nu_2\nu_1}_{\substack{\mathbf{ji},\mu_1\mu_2 \\ I_eI_h}} \right|^2 \delta\big(E_\mathbf{i}^{I_eI_h} - E_\mathbf{j}^{I_e+1I_h(+1)} + \epsilon_{\nu_1}^{\mu_1} + \epsilon_{\nu_2}^{\mu_2} - \epsilon_{\nu_3}^{\mu_2}\big) \big(f_{\nu_3}^{\mu_2}\big(1-f_{\nu_2}^{\mu_2}\big)\big(1-f_{\nu_1}^{\mu_1}\big)\rho_\mathbf{j}^{I_e+1I_h(+1)} - \big(1-f_{\nu_3}^{\mu_2}\big) f_{\nu_2}^{\mu_2} f_{\nu_1}^{\mu_1} \rho_\mathbf{i}^{I_eI_h}\big), \tag{18}$$

or within the QD:

$$T^{\nu_1}_{\substack{\mathbf{ij},\mu_1\mu_2 \\ I_e+1;\, I_h}} = \left| \tilde{V}^{\nu_1}_{\substack{\mathbf{ji},\mu_1\mu_2 \\ I_eI_h}} \right|^2 \delta\big(E_\mathbf{i}^{I_eI_h} - E_\mathbf{j}^{I_e+1I_h} + \epsilon_{\nu_1}^{\mu_1}\big) \times \big(\big(1-f_{\nu_1}^{\mu_1}\big)\rho_\mathbf{j}^{I_e+1I_h} - f_{\nu_1}^{\mu_1}\rho_\mathbf{i}^{I_eI_h}\big). \tag{19}$$

In principle, both Coulomb scatterings depend on similar contributions: the squared effective Coulomb coupling element, the energy conserving δ-function including continuum energies ϵ_ν^μ, and the eigenenergies $E_\mathbf{i}^{I_eI_h}$ of the QD, typical Boltzmann-type structures and density matrix contributions for the occupation probability of QD configurations.

Usually, reservoir assisted scattering Eq. (18) constitutes the dominant contribution, since the energy conservation during the scattering process can be achieved much easier in the continuum Eq. (18) than in the QD Eq. (19), cf. Fig. 2. In case of phonon assisted processes an additional energy is available to achieve the energy conservation for QD assisted scattering.

The first (second) term in Eqs. (18) and (19) describes QD out- (in-) scattering, including the Pauli blocking of the final reservoir occupations after scattering, e.g., $(1-f_{\nu_1}^{\mu_1})$. Scattering contributions in Boltzmann equations for electrons in a continuum typically have the form $f_1 f_2(1-f_3)(1-f_4)$. Beside Boltzmann like treatment of carrier scattering processes in continuum systems scattering in systems with discrete levels like molecules, atoms or QDs can be described via density matrix equations including $\Gamma_{\mathbf{j}\to\mathbf{i}}\rho_\mathbf{j}^{I_e+1I_h} - \Gamma_{\mathbf{i}\to\mathbf{j}}\rho_\mathbf{i}^{I_eI_h}$. In contrast to pure Boltzmann and pure density matrix equations in the approach developed here a hybrid form enters like $f_{\nu_3}^{\mu_2}(1-f_{\nu_2}^{\mu_2})(1-f_{\nu_1}^{\mu_1})\rho_\mathbf{j}^{I_e+1I_h}$ [cf. Eq. (18) first term], showing the full strength of the hybrid density matrix approach. The scattering processes changes the occupation probability of the QD configuration $\rho_\mathbf{i}^{I_eI_h}$ to the QD configuration $\rho_\mathbf{j}^{I_e+1I_h}$ executing at the same time a Pauli blocking process in the continuum.

For the case of electron-electron (or hole-hole) scattering including carrier relaxation within the reservoir, the carriers in the reservoir are indistinguishable particles. This results in interference effects similar to double slit experiments [43]. In the equations of motion, those interference effects are indicated by differences of Coulomb coupling elements like $\tilde{V}^{\nu_3\nu_2\nu_1}_{\substack{\mathbf{ji},ee \\ I_eI_h}} - \tilde{V}^{\nu_3\nu_1\nu_2}_{\substack{\mathbf{ji},ee \\ I_eI_h}}$, cf. Appendix.[4]

VI. GENERAL DISCUSSION

A well-known limit can be observed for the hybrid density matrix equations, if we assume an quasiequilibrium in the reservoir. In this case, the equation of motion for the QD density matrix resembles the typical Lindblad form [28,32] for QD reduced density matrix, cf. Eqs. (16) and (17). However, the hybrid density matrix approach is able to treat the continuum dynamically Eq. (17) and to go beyond the Markovian treatment of the Lindblad coupling, cf. Appendix. In particular, this is necessary to describe the dynamics of the time modulated reservoir in electrically pumped QD devices.

In addition, to discuss the potential as well as the intrinsic limits of nonclassical light emission from QD devices, the sequence of electrical generation, time evolution and subsequent radiative decay from different excited many particle configurations in QD devices must be investigated. The presented method offers the possibility to describe effects of generation, propagation and radiative decay of the many body configurations.

Furthermore, the presented hybrid density matrix approach results in a nonlinear, closed set of differential equations, c.f. Appendix, Eqs. (A1)–(A5), which for example can be numerically solved using Runge-Kutta or other time solving methods. The most demanding aspect in the numerical implementation of our equations is the sum over a sufficient number of continuum carrier states for achieving numerical convergence. Obviously, the solution of the hybrid equations require knowledge of the coupling elements (in the QD example the Coulomb coupling elements). For energetically higher continuum levels, the coupling to the systems with discrete resonances decreases at some point. Furthermore, the continuum carrier occupations are only significantly filled near the continuum minimum (band edge). Both points can be used to achieve numerical convergence, including only a finite number of continuum states in numerical simulations.

We discuss the specific example of a QD-continuum system with respect to numerical feasibility: here, the calculation of Coulomb coupling elements is quite challenging due to the number of integrals involved in the two particle interaction, which must be calculated for the continuum states. As an overall estimation, the numerical effort to solve the set of hybrid equations should be manageable similarly to Ref. [44].

[3]The occurring difference of Coulomb coupling elements $\tilde{V}^{\nu_2\nu_1\nu}_{\mathbf{ji},ee} - \tilde{V}^{\nu_2\nu\nu_1}_{\mathbf{ji},ee}$ is redefined to $\tilde{V}^{\nu_2\nu_1\nu}_{\mathbf{ji},ee}$.

[4]For the configuration inside the QD, interference effects are already included during diagonalization.

With high numerical effort, interesting results of the switch-on dynamics of the wetting layer and QD populations can be obtained at least in the Markov approximation as in Ref. [44]. In comparison to Ref. [44], the main difference is that the hybrid equations will allow to include the influence of electron hole complexes like excitons, trions, biexcitons, etc., with systematically including all their intradot correlations in the QD with similar order of numerical effort as in Ref. [44], which did not include complexes. The inclusion of complexes like biexcitons [28,29] is particularly important in nitrides, since interesting bright and dark biexciton configurations are expected [45].

In standard correlation expansion as in Ref. [44] no systematic way exists to include only the intra dot complexes. Their implementation would result in a much higher numerical effort in pure correlation expansion. Additionally, in a non-Markovian evaluation, the time dependence of scattering channels from continuum to discrete states (i.e., the complexes like excitons or biexcitons in QDs), as it was, e.g., seen in bound systems [42,46], is an expected feature.

VII. CONCLUSION

In conclusion, we present a hybrid density matrix approach, which combines conventional correlation expansion with an exact diagonalization scheme in a projector operator technique to describe hybrid systems (e.g., QD reservoir). In the limit of an equilibrated reservoir, the resulting hybrid density matrix equations give well known results. However, they offer a non-Markovian description, because the influence of the reservoir is not restricted to a treatment via Lindblad terms.

Beside Coulomb coupling, further interactions can be included such as non-Markovian, nonperturbative electron-phonon interaction [35,47] via self-consistent Born or time-convolutionless (TCL) [48] approximations, in particular, to discuss the zero phonon line broadening or electron-phonon induced spin-flips, which is expected to be relevant for nitrides QDs because of their enhanced electron-phonon coupling strength.

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APPENDIX: NON-MARKOVIAN SET OF EQUATION OF MOTION

The equation of motion of the reduced density matrix of the QD beyond the Markov approximation read

$$\begin{aligned}\partial_t \rho_{\mathbf{i}}^{I_e I_h} =& -\frac{2\xi_e}{\hbar}\mathrm{Im}\left\{\sum_{\mathbf{j},\nu_1}\left[\left(\tilde{V}^{\nu_1}_{\substack{\mathbf{ij},ee\\ I_eI_h}}+\tilde{V}^{\nu_1}_{\substack{\mathbf{ij},eh\\ I_eI_h}}\right)^*\left\langle X_{\mathbf{i}}^{I_eI_h\dagger}e_{\nu_1}^\dagger X_{\mathbf{j}}^{I_e+1I_h}\right\rangle-\left(\tilde{V}^{\nu_1}_{\substack{\mathbf{ji},eh\\ I_e-1I_h}}+\tilde{V}^{\nu_1}_{\substack{\mathbf{ji},ee\\ I_e-1I_h}}\right)^*\left\langle X_{\mathbf{j}}^{I_e-1I_h\dagger}e_{\nu_1}^\dagger X_{\mathbf{i}}^{I_eI_h}\right\rangle\right.\\ &+\sum_{\nu_2,\nu_3}\left(\tilde{V}^{\nu_3\nu_2\nu_1}_{\substack{\mathbf{ij},ee\\ I_eI_h}}{}^*\left\langle X_{\mathbf{i}}^{I_eI_h\dagger}e_{\nu_1}^\dagger e_{\nu_2}^\dagger e_{\nu_3}X_{\mathbf{j}}^{I_e+1I_h}\right\rangle-\tilde{V}^{\nu_3\nu_2\nu_1}_{\substack{\mathbf{ji},ee\\ I_e-1I_h}}{}^*\left\langle X_{\mathbf{j}}^{I_e-1I_h\dagger}e_{\nu_1}^\dagger e_{\nu_2}^\dagger e_{\nu_3}X_{\mathbf{i}}^{I_eI_h}\right\rangle\right.\\ &\left.\left.\left.+\tilde{V}^{\nu_1\nu_3\nu_2}_{\substack{\mathbf{ij},eh\\ I_eI_h}}{}^*\left\langle X_{\mathbf{i}}^{I_eI_h\dagger}e_{\nu_1}^\dagger h_{\nu_2}^\dagger h_{\nu_3}X_{\mathbf{j}}^{I_e+1I_h}\right\rangle-\tilde{V}^{\nu_1\nu_3\nu_2}_{\substack{\mathbf{ji},eh\\ I_e-1I_h}}{}^*\left\langle X_{\mathbf{j}}^{I_e-1I_h\dagger}e_{\nu_1}^\dagger h_{\nu_2}^\dagger h_{\nu_3}X_{\mathbf{i}}^{I_eI_h}\right\rangle\right)\right]\right\}+e\leftrightarrow h,\end{aligned}\tag{A1}$$

where $e\leftrightarrow h$ means, that all listed contributions enter again with exchanged carrier indices e and h. The average populations in the reservoir as a new dynamical variable is given by

$$\begin{aligned}\partial_t f_\nu^e=&-\frac{2}{\hbar}\mathrm{Im}\left\{\sum_{I_e,I_h,\mathbf{i},\mathbf{j}}\left(\xi_e\tilde{V}^{\nu}_{\substack{\mathbf{ji},ee\\ I_e+1I_h}}{}^*\left\langle X_{\mathbf{i}}^{I_e+1I_h\dagger}e_\nu^\dagger X_{\mathbf{j}}^{I_e+2I_h}\right\rangle+\xi_e\tilde{V}^{\nu}_{\substack{\mathbf{ji},eh\\ I_eI_h+1}}{}^*\left\langle X_{\mathbf{i}}^{I_eI_h+1\dagger}e_\nu^\dagger X_{\mathbf{j}}^{I_e+1I_h+1}\right\rangle\right.\right.\\ &+\sum_{\nu_1,\nu_2}\left[\xi_e\left(\tilde{V}^{\nu_2\nu\nu_1}_{\substack{\mathbf{ji},ee\\ I_eI_h}}-\tilde{V}^{\nu_2\nu_1\nu}_{\substack{\mathbf{ji},ee\\ I_eI_h}}\right)^*\left\langle X_{\mathbf{i}}^{I_eI_h\dagger}e_\nu^\dagger e_{\nu_1}^\dagger e_{\nu_2}X_{\mathbf{j}}^{I_e+1I_h}\right\rangle+\xi_e\tilde{V}^{\nu\nu_2\nu_1}_{\substack{\mathbf{ji},ee\\ I_eI_h}}{}^*\left\langle X_{\mathbf{i}}^{I_eI_h\dagger}e_{\nu_1}^\dagger e_{\nu_2}^\dagger e_\nu X_{\mathbf{j}}^{I_e+1I_h}\right\rangle\right.\\ &\left.\left.\left.-\xi_e\tilde{V}^{\nu_2\nu_1\nu}_{\substack{\mathbf{ji},eh\\ I_eI_h}}{}^*\left\langle X_{\mathbf{i}}^{I_eI_h\dagger}e_\nu^\dagger h_{\nu_1}^\dagger h_{\nu_2}X_{\mathbf{j}}^{I_e+1I_h}\right\rangle-\xi_h\tilde{V}^{\nu\nu_1\nu_2}_{\substack{\mathbf{ji},he\\ I_eI_h}}{}^*\left\langle X_{\mathbf{i}}^{I_eI_h\dagger}e_{\nu_1}^\dagger h_{\nu_2}^\dagger e_\nu X_{\mathbf{j}}^{I_eI_h+1}\right\rangle+\xi_h\tilde{V}^{\nu_2\nu\nu_1}_{\substack{\mathbf{ji},he\\ I_eI_h}}{}^*\left\langle X_{\mathbf{i}}^{I_eI_h\dagger}e_\nu^\dagger h_{\nu_1}^\dagger e_{\nu_2}X_{\mathbf{j}}^{I_eI_h+1}\right\rangle\right)\right]\right\}\end{aligned}\tag{A2}$$

with $\xi_e=(-1)^{I_e+I_h}$ and $\xi_h=(-1)^{I_h}$. The hole continuum can be described in an identical way compared to the electron continuum. QD as well as continuum assisted electron-electron, hole-hole, electron-hole, and hole-electron scattering occurs in Eqs. (A1) and (A2), cf. Fig. 2.

The system observables $\rho_{\mathbf{i}}^{I_e I_h}$ and f_ν^μ couple to the non-Markovian equations of mixed expectation values. Carrier capture processes including carrier relaxation within the QD are represented by

$$
\begin{aligned}
&-i\hbar\partial_t\left\langle X_{\mathbf{i}}^{I_e I_h\dagger} e_\nu^\dagger X_{\mathbf{j}}^{I_e+1 I_h}\right\rangle = \left(E_{\mathbf{i}}^{I_e I_h} - E_{\mathbf{j}}^{I_e+1 I_h} + \epsilon_\nu^c\right)\left\langle X_{\mathbf{i}}^{I_e I_h\dagger} e_\nu^\dagger X_{\mathbf{j}}^{I_e+1 I_h}\right\rangle \\
&+\xi_e\left[\tilde{V}^{\ \nu}_{\substack{\mathbf{ji},ee\\ I_e I_h}} + \tilde{V}^{\ \nu}_{\substack{\mathbf{ji},eh\\ I_e I_h}} + \sum_{\nu_1}\left(\tilde{V}^{\ \nu_1\nu_1\nu}_{\substack{\mathbf{ji},ee\\ I_e I_h}} - \tilde{V}^{\ \nu_1\nu\nu_1}_{\substack{\mathbf{ji},ee\\ I_e I_h}}\right) f_{\nu_1}^e\right]\Gamma^{\ \nu}_{\substack{\mathbf{ij},e\\ I_e+1;\, I_h}} + \xi_e\sum_{\nu_1}\tilde{V}^{\ \nu_1\nu_1\nu}_{\substack{\mathbf{ji},eh\\ I_e I_h}} f_\nu^e\Gamma^{\ \nu_1}_{\substack{\mathbf{ij},h\\ I_e+1;\, I_h}},
\end{aligned}
\tag{A3}
$$

where we introduce the definition $\Gamma^{\ \nu_1}_{\substack{\mathbf{ij},\mu_1\\ I_e+1;\, I_h}} = (1-f_{\nu_1}^{\mu_1})\rho_{\mathbf{j}}^{I_e+1 I_h} - f_{\nu_1}^{\mu_1}\rho_{\mathbf{i}}^{I_e I_h}$, in which the first contribution represents the QD out- and the second the QD in-scattering. In the corresponding hole quantity, the carrier index is changed. Additionally, scattering processes containing carrier relaxation within the reservoir enter, such as electron-electron scattering:

$$
\begin{aligned}
&-i\hbar\partial_t\left\langle X_{\mathbf{i}}^{I_e I_h\dagger} e_{\nu_1}^\dagger e_{\nu_2}^\dagger e_{\nu_3} X_{\mathbf{j}}^{I_e+1 I_h}\right\rangle = \left(E_{\mathbf{i}}^{I_e I_h} - E_{\mathbf{j}}^{I_e+1 I_h} + \epsilon_{\nu_1}^c + \epsilon_{\nu_2}^c - \epsilon_{\nu_3}^c\right)\left\langle X_{\mathbf{i}}^{I_e I_h\dagger} e_{\nu_1}^\dagger e_{\nu_2}^\dagger e_{\nu_3} X_{\mathbf{j}}^{I_e+1 I_h}\right\rangle \\
&+\xi_e\left(\tilde{V}^{\ \nu_3\nu_2\nu_1}_{\substack{\mathbf{ji},ee\\ I_e I_h}} - \tilde{V}^{\ \nu_3\nu_1\nu_2}_{\substack{\mathbf{ji},ee\\ I_e I_h}}\right)\left[f_{\nu_3}^e\left(1-f_{\nu_2}^e\right)\left(1-f_{\nu_1}^e\right)\rho_{\mathbf{j}}^{I_e+1 I_h} - \left(1-f_{\nu_3}^e\right) f_{\nu_2}^e f_{\nu_1}^e\rho_{\mathbf{i}}^{I_e I_h}\right] \\
&-\xi_e\delta_{\nu_1,\nu_3} f_{\nu_1}^e\left[\sum_{\nu_4}\left(\tilde{V}^{\ \nu_4\nu_4\nu_2}_{\substack{\mathbf{ji},ee\\ I_e I_h}} - \tilde{V}^{\ \nu_4\nu_2\nu_4}_{\substack{\mathbf{ji},ee\\ I_e I_h}}\right) f_{\nu_4}^e + \sum_{\nu_4}\tilde{V}^{\ \nu_4\nu_4\nu_2}_{\substack{\mathbf{ji},eh\\ I_e I_h}} f_{\nu_4}^h - \tilde{V}^{\ \nu_2}_{\substack{\mathbf{ji},ee\\ I_e I_h}} - \tilde{V}^{\ \nu_2}_{\substack{\mathbf{ji},eh\\ I_e I_h}}\right]\Gamma^{\ \nu_2}_{\substack{\mathbf{ij},e\\ I_e+1;\, I_h}} \\
&+\xi_e\delta_{\nu_2,\nu_3} f_{\nu_2}^e\left[\sum_{\nu_4}\left(\tilde{V}^{\ \nu_4\nu_4\nu_1}_{\substack{\mathbf{ji},ee\\ I_e I_h}} - \tilde{V}^{\ \nu_4\nu_1\nu_4}_{\substack{\mathbf{ji},ee\\ I_e I_h}}\right) f_{\nu_4}^e + \sum_{\nu_4}\tilde{V}^{\ \nu_4\nu_4\nu_1}_{\substack{\mathbf{ji},eh\\ I_e I_h}} f_{\nu_4}^h - \tilde{V}^{\ \nu_1}_{\substack{\mathbf{ji},ee\\ I_e I_h}} - \tilde{V}^{\ \nu_1}_{\substack{\mathbf{ji},eh\\ I_e I_h}}\right]\Gamma^{\ \nu_1}_{\substack{\mathbf{ij},ee\\ I_e+1;\, I_h}},
\end{aligned}
\tag{A4}
$$

or electron-hole scattering:

$$
\begin{aligned}
&-i\hbar\partial_t\left\langle X_{\mathbf{i}}^{I_e I_h\dagger} e_{\nu_1}^\dagger h_{\nu_2}^\dagger e_{\nu_3} X_{\mathbf{j}}^{I_e I_h+1}\right\rangle = \left(E_{\mathbf{i}}^{I_e I_h} - E_{\mathbf{j}}^{I_e I_h+1} + \epsilon_{\nu_1}^c + \epsilon_{\nu_2}^v - \epsilon_{\nu_3}^c\right)\left\langle X_{\mathbf{i}}^{I_e I_h\dagger} e_{\nu_1}^\dagger h_{\nu_2}^\dagger e_{\nu_3} X_{\mathbf{j}}^{I_e I_h+1}\right\rangle \\
&-\xi_h\tilde{V}^{\ \nu_1\nu_3\nu_2}_{\substack{\mathbf{ji},eh\\ I_e I_h}}\left[f_{\nu_3}^e\left(1-f_{\nu_2}^h\right)\left(1-f_{\nu_1}^e\right)\rho_{\mathbf{j}}^{I_e I_h+1} - \left(1-f_{\nu_3}^e\right) f_{\nu_2}^h f_{\nu_1}^e\rho_{\mathbf{i}}^{I_e I_h}\right] \\
&-\xi_h\delta_{\nu_1,\nu_3} f_{\nu_1}^e\left[\sum_{\nu_4}\left(\tilde{V}^{\ \nu_4\nu_4\nu_2}_{\substack{\mathbf{ji},hh\\ I_e I_h}} - \tilde{V}^{\ \nu_4\nu_2\nu_4}_{\substack{\mathbf{ji},hh\\ I_e I_h}}\right) f_{\nu_4}^h + \sum_{\nu_4}\tilde{V}^{\ \nu_4\nu_4\nu_2}_{\substack{\mathbf{ji},eh\\ I_e I_h}} f_{\nu_4}^e - \tilde{V}^{\ \nu_2}_{\substack{\mathbf{ji},hh\\ I_e I_h}} - \tilde{V}^{\ \nu_2}_{\substack{\mathbf{ji},eh\\ I_e I_h}}\right]\Gamma^{\ \nu_2}_{\substack{\mathbf{ij},h\\ I_e+1;\, I_h}}.
\end{aligned}
\tag{A5}
$$

Again, the carrier index must be changed to describe the inverse process regarding electrons and holes. Additional non-Markovian contributions occur, if the continuum indices of creation and annihilation electron operators in Eqs. (A4) and (A5) are identical. In the Markovian limit, these terms do not enter in the dynamics due to the energy conservation. However, they could play a role in higher-order Markov treatment, resulting in Coulomb induced dynamical damping or energy renormalization.

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