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Quantum transport theory for semiconductor nanostructures: A density-matrix formulation

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A general density-matrix formulation of quantum transport phenomena in semiconductor nanostructures is presented. More specifically, contrary to the conventional single-particle correlation expansion, we shall investigate separately the effects of the adiabatic or Markov limit and of the reduction procedure. Our fully operatorial approach allows us to better identify the general properties of the scattering superoperators entering our effective quantum transport theory at various description levels, e.g., N electrons-plus-quasiparticles, N electrons only, and single-particle picture. In addition to coherent transport phenomena characterizing the transient response of the system, the proposed theoretical description allows us to study scattering induced phase coherence in steady-state conditions. As a prototypical example, we shall investigate polaronic effects in strongly biased semiconductor superlattices.

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I. INTRODUCTION

Recent developments in nanoscience/technology push device miniaturization toward limits where the traditional semiclassical transport treatments[1] can no longer be employed, and more rigorous quantum transport approaches are imperative.[2] However, in spite of the quantum-mechanical nature of carrier dynamics in the core region of typical nanostructured devices—like semiconductor superlattices, double-barrier structures, and quantum dots—the overall behavior of such quantum systems is often the result of a nontrivial interplay between phase coherence and energy relaxation/dephasing.[3] It follows that a proper treatment of such novel nanoscale devices requires a theoretical modeling able to properly account for both coherent and incoherent, i.e., phase-breaking, processes on the same footing within a many-body picture.

More precisely, the idealized behavior of a so-called "quantum device"[4] is usually described via the elementary physical picture of the square-well potential and/or in terms of a simple quantum-mechanical n-level system. For a quantitative investigation of state-of-the-art quantum optoelectronic devices, however, two features strongly influence and modify such a simplified scenario: (i) the intrinsic many-body nature of the carrier system under investigation, and (ii) the potential coupling of the electronic subsystem of interest with a variety of interaction mechanisms, including the presence of spatial boundaries.[5–7]

The wide family of so-called quantum devices can be divided into two main classes: the first one grouping semiconductor devices characterized by a genuine quantum-mechanical behavior of their carrier subsystem, and a second one which comprises low-dimensional nanostructures whose transport dynamics may be safely treated within the semiclassical picture.

Devices within the first class—characterized by a weak coupling of the carrier subsystem with the host material—are natural candidates for the implementation of quantum information/computation processing.[8] These include, in particular, semiconductor quantum-dot structures,[9] for which all-optical implementations have been recently proposed.[10–12] In this case, the pure quantum-mechanical carrier dynamics is only weakly disturbed by decoherence processes; therefore, the latter are usually described in terms of extremely simplified models.

Conversely, quantum devices in the second class—in spite of their partially discrete energy spectrum due to spatial carrier confinement—exhibit a carrier dynamics which can be still described via a semiclassical scattering picture. Such optoelectronic nanostructured devices include multi-quantum-well and superlattice structures, like quantum-cascade lasers (QCLs).[13,14] These systems are characterized by a strong interplay between coherent dynamics and energy-relaxation/dephasing processes; it follows that for a quantitative description of such nontrivial coherence/dissipation coupling the latter need to be treated via fully microscopic models.

In this paper we shall primarily focus on this second class of quantum devices, providing a comprehensive microscopic theory of charge transport in semiconductor nanostructures based on the well-known density-matrix approach. It is worth mentioning that an alternative approach, equivalent to the density-matrix formalism employed in this paper, is given by the nonequilibrium Green's function technique; the latter can be regarded as an extension of the well-known equilibrium or zero-temperature Green's function theory to nonequilibrium regimes, introduced in the 1960s by Kadanoff and Baym[15] and Keldysh.[16] An introduction to the theory of nonequilibrium Green's functions with applications to many problems in transport and optics of semiconductors can be found in the book by Haug and Jauho.[17] By employing—and further developing and extending—such nonequilibrium Green's function formalism, a number of groups have proposed efficient quantum-transport treatments for the study of various semiconductor nanostructures[18] as well as of modern micro/optoelectronic devices.[19]

Within the general density-matrix formalism two different strategies are commonly employed: (i) the quantum-kinetic

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treatment,[3] and (ii) the description based on the Liouville-von Neumann equation.[20]

The primary goal of a quantum-kinetic theory is to evaluate the temporal evolution of a reduced set of single- or few-particle quantities directly related to the electro-optical phenomenon under investigation, the so-called kinetic variables of the system. However, due to the many-body nature of the problem, an exact solution in general is not possible; it follows that for a detailed understanding realistic semiconductor models have to be considered, which then can only be treated approximately. Within the kinetic-theory approach one starts directly with the equations of motion for the single-particle density matrix. Due to the many-body nature of the problem, the resulting set of equations of motion is not closed; instead, it constitutes the starting point of an infinite hierarchy of higher-order density matrices. Besides differences related to the quantum statistics of the quasiparticles involved, this is equivalent to the Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy in classical gas dynamics.[21] The central approximation in this formalism is the truncation of the hierarchy. This can be based on different physical pictures. A common approach is to use the argument that correlations involving an increasing number of particles will become less and less important.[3] An alternative quantum-kinetic scheme—based on an expansion in powers of the exciting laser field—has been introduced by Axt and Stahl, the so-called "dynamics controlled truncation" (DCT).[22]

Within the treatment based on the Liouville-von Neumann equation, the starting point is the equation of motion for the global density-matrix operator, describing many electron plus various quasiparticle excitations. The physical quantities of interest for the electronic subsystem are then typically derived via a suitable "reduction procedure," aimed at tracing out nonrelevant degrees of freedom. Contrary to the kinetic theory, this approach has allowed for a fully quantum-mechanical treatment of high-field transport in semiconductors,[23] thus overcoming some of the basic limitations of conventional kinetic treatments, e.g., the completed-collision limit and the Markov approximation.

The primary goal of the present paper is to discuss in very general terms the physical properties and validity limits of the so-called "adiabatic" or Markov approximation. Within the traditional semiclassical or Boltzmann theory, this approximation is typically introduced together with the so-called diagonal approximation, i.e., the neglect of nondiagonal density-matrix elements. However, as described in Ref. 24, the Markov limit can also be performed within a fully nondiagonal density-matrix treatment of the problem; this leads to the introduction of generalized in- and out-scattering superoperators, whose general properties and physical interpretation are not straightforward. In particular, it is imperative to understand if—and under which conditions—the adiabatic or Markov approximation preserves the positive-definite character of our reduced density matrix; indeed, this distinguished property is generally lost within the quantum-kinetic approaches previously mentioned.[3,24] To this end, starting from the Liouville-von Neumann-equation approach, we shall propose a very general treatment of the Markov approximation. More specifically, contrary to the conventional single-particle correlation expansion of the kinetic theory, we shall investigate separately the effects of the Markov limit and of the reduction procedure. Our fully operatorial approach will allow us to better identify the general properties of the scattering superoperators entering our effective quantum transport theory at various description levels, e.g., N electrons-plus-quasiparticles, N electrons only, and single-particle picture. In addition to coherent transport phenomena characterizing the transient response of the system, the proposed theoretical description allows us to study scattering induced phase coherence in steady-state conditions. In particular, based on the proposed approach we shall consider—as a prototypical example—polaronic coherence in strongly biased semiconductor superlattices.

The paper is organized as follows. In Sec. II we present the proposed theoretical approach: after specifying the physical system under investigation (Sec. II A) and recalling the fundamentals of the density-matrix formalism (Sec. II B), we shall introduce the Markov approximation (Sec. II C) and derive the explicit form of the scattering superoperators (Sec. II D) as well as their semiclassical counterparts (Sec. II E); we shall then discuss the so-called reduction procedure (Sec. II F) and the single-particle description (Sec. II G) for carrier-carrier as well as carrier-quasiparticle interactions. In Sec. III we shall address the general problem of scattering-induced phase coherence; in particular, we shall present a few simulated experiments concerning polaronic coherence in semiconductor superlattices. Finally, in Sec. V we shall summarize and draw some conclusions.

II. THEORETICAL BACKGROUND

A. Physical system

To provide a general formulation of quantum charge transport in semiconductor nanostructures, let us consider a generic carrier gas within a semiconductor crystal in the presence of electromagnetic fields. The corresponding Hamiltonian can be schematically written as

$$\hat{H} = \hat{H}_{\mathrm{o}} + \hat{H}'. \tag{1}$$

The first term,

$$\hat{H}_{\mathrm{o}} = \hat{H}_{\mathrm{o}}^{c} + \hat{H}_{\mathrm{o}}^{qp} = \sum_{\alpha} \epsilon_{\alpha} \hat{c}_{\alpha}^{\dagger} \hat{c}_{\alpha} + \sum_{\mathbf{q}} \epsilon_{\mathbf{q}} \hat{b}_{\mathbf{q}}^{\dagger} \hat{b}_{\mathbf{q}}, \tag{2}$$

is the sum of the free-carrier and free-quasiparticle Hamiltonians, where the Fermionic operators $\hat{c}_{\alpha}^{\dagger}$ ($\hat{c}_{\alpha}$) denote creation (destruction) of a carrier in the single-particle state α (with energy ϵ_{α}), while the Bosonic operators $\hat{b}_{\mathbf{q}}^{\dagger}$ ($\hat{b}_{\mathbf{q}}$) denote creation (destruction) of a generic quasiparticle excitation with wave vector $\mathbf{q}$ and energy $\epsilon_{\mathbf{q}}$, i.e., phonons, photons, plasmons, etc.

The second term, $\hat{H}'$, is the sum of all possible carrier-carrier as well as carrier-quasiparticle interaction Hamiltonians, i.e., carrier-phonon, carrier-photon, carrier-plasmon, etc.

The noninteracting carrier-plus-quasiparticle basis states are given by the eigenstates of $\hat{H}_{\mathrm{o}}$: the generic eigenstate

$|\lambda\rangle = |\{n_\alpha\}\rangle \otimes |\{n_{\mathbf{q}}\}\rangle$ is the tensor product of noninteracting carrier and quasiparticle states corresponding, respectively, to the occupation numbers $\{n_\alpha\}$ and $\{n_{\mathbf{q}}\}$, while the noninteracting energy spectrum $\epsilon_\lambda = \Sigma_\alpha \epsilon_\alpha n_\alpha + \Sigma_{\mathbf{q}} \epsilon_{\mathbf{q}} n_{\mathbf{q}}$ is the sum of the total carrier and quasiparticle energies.

The interaction Hamiltonian $\hat{H}'$ cannot in general be treated exactly. A typical approach consists of regarding it as a perturbation acting on the noninteracting carrier-plus-quasiparticle states $\{|\lambda\rangle\}$. In this context, the basic ingredients are the matrix elements of $\hat{H}'$ within our noninteracting basis states: $H'_{\lambda\lambda'} = \langle\lambda|\hat{H}'|\lambda'\rangle$.

B. Density-matrix formalism

In view of the huge number of degrees of freedom $(\{\alpha\},\{\mathbf{q}\})$ involved in the microscopic treatment of any solid-state system, a statistical description of the problem is imperative. As we shall see (in Sec. II F), this will result in a suitable statistical average over "nonrelevant" degrees of freedom.

Given a physical quantity A—described by the operator $\hat{A}$—its quantum plus statistical average value is given by

$$A = \overline{\langle\psi|\hat{A}|\psi\rangle} = \mathrm{tr}\{\hat{A}\hat{\rho}\}, \tag{3}$$

where

$$\hat{\rho} = \overline{|\psi\rangle\langle\psi|} \tag{4}$$

is the so-called density-matrix operator. The latter is defined as the statistical average of the projection operator corresponding to the generic state vector $|\psi\rangle$ of the system, and can then be regarded as the statistical generalization of the quantum-mechanical concept of state vector.

Starting from the global Schrödinger equation describing our interacting carrier-plus-quasiparticle many-body system, the following Liouville-von Neumann equation of motion for the density-matrix operator can be readily obtained:

$$\frac{d\hat{\rho}}{dt} = \mathcal{L}(\hat{\rho}) = \frac{1}{i\hbar}[\hat{H},\hat{\rho}], \tag{5}$$

where $\mathcal{L}$ is usually referred to as Liouville superoperator. Equation (5) can be regarded as the statistical generalization of the Schrödinger equation; its exact solution is given by

$$\hat{\rho}(t) = e^{\mathcal{L}(t-t_0)}\hat{\rho}(t_0) = \hat{U}(t-t_0)\hat{\rho}(t_0)\hat{U}^\dagger(t-t_0), \tag{6}$$

where

$$\hat{U}(t-t_0) = e^{\hat{H}(t-t_0)/i\hbar} \tag{7}$$

is the evolution operator corresponding to the total Hamiltonian $\hat{H}$ in Eq. (1). Such an exact solution corresponds to a fully quantum-mechanical unitary evolution of the whole many-body system, i.e., no energy relaxation/dephasing. Indeed, it is easy to verify that the total quantum entropy

$$S = -k_{\mathrm{B}}\,\mathrm{tr}\{\hat{\rho}\log\hat{\rho}\} \tag{8}$$

is not affected by the unitary transformation in Eq. (7).[25]

As anticipated, the total many-body Hamiltonian in Eq. (1) cannot be treated exactly. The aim of a quantum-transport theory is to derive effective equations describing the carrier subsystem of interest within some approximation scheme; this is typically realized via the following two basic steps: first an adiabatic decoupling between the different time scales induced by $\hat{H}_{\mathrm{o}}$ and $\hat{H}'$—called Markov limit—and then a projection of the global system dynamics over a subsystem of interest via the introduction of a so-called reduced density-matrix operator.

C. The adiabatic or Markov approximation

Starting from the separation $\hat{H} = \hat{H}_{\mathrm{o}} + \hat{H}'$ in Eq. (1), the Liouville-von Neumann equation (5) can be written as

$$\frac{d\hat{\rho}}{dt} = \left.\frac{d\hat{\rho}}{dt}\right|_{\hat{H}_{\mathrm{o}}} + \left.\frac{d\hat{\rho}}{dt}\right|_{\hat{H}'}, \tag{9}$$

where the two contributions describe, respectively, the time evolution induced by the noninteracting Hamiltonian $\hat{H}_{\mathrm{o}}$ and the interaction term $\hat{H}'$.

The first contribution can be treated exactly within the standard interaction scheme. Indeed, it is easy to show that the time evolution of the density-matrix operator in the interaction picture,

$$\hat{\rho}^i = \hat{U}_{\mathrm{o}}^\dagger(t-t_0)\hat{\rho}\hat{U}_{\mathrm{o}}(t-t_0), \tag{10}$$

is simply given by

$$\frac{d\hat{\rho}^i}{dt} = -i[\hat{\mathcal{H}}^i,\hat{\rho}^i], \tag{11}$$

where $\hat{U}_{\mathrm{o}}(t-t_0)$ is the evolution operator corresponding to the noninteracting Hamiltonian $\hat{H}_{\mathrm{o}}$, and

$$\hat{\mathcal{H}}^i(t) = \hat{U}_{\mathrm{o}}^\dagger(t-t_0)\frac{\hat{H}'}{\hbar}\hat{U}_{\mathrm{o}}(t-t_0) \tag{12}$$

denotes the Hamiltonian $\hat{H}'$ in units of $\hbar$ within the interaction picture.

The key idea beyond any perturbation approach is that the effect of the interaction Hamiltonian $\hat{H}'$ is "small" compared to the free evolution dictated by the noninteracting Hamiltonian $\hat{H}_{\mathrm{o}}$. More precisely, the interaction matrix elements $H'_{\lambda\lambda'}$ are smaller than the typical energy difference $\epsilon_\lambda - \epsilon_{\lambda'}$.

Following this spirit, by formally integrating Eq. (11) from t_0 to the current time t, we get

$$\hat{\rho}^i(t) = \hat{\rho}^i(t_0) - i\int_{t_0}^{t} dt'[\hat{\mathcal{H}}^i(t'),\hat{\rho}^i(t')]. \tag{13}$$

By inserting the above formal solution for $\hat{\rho}^i(t)$ on the right-hand side of Eq. (11) we obtain an integro-differential equation of the form

$$\frac{d}{dt}\hat{\rho}^i(t) = -i[\hat{\mathcal{H}}^i(t),\hat{\rho}^i(t_0)] - \int_{t_0}^{t} dt'[\hat{\mathcal{H}}^i(t),[\hat{\mathcal{H}}^i(t'),\hat{\rho}^i(t')]]. \tag{14}$$

We stress that so far no approximation has been introduced: Equations (11), (13), and (14) are all fully equivalent, we have just isolated the first-order contribution from the full time evolution in Eq. (11). It is then clear that, by iteratively substituting Eq. (13) into itself, the above procedure can be extended to any perturbation order. This leads to the well-known Neuman series:

$$\hat{\rho}^i(t) = \hat{\rho}^i(t_0) + \sum_{n=1}^{\infty}(-i)^n \int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 \cdots \int_{t_0}^{t_{n-1}} dt_n \times[\hat{\mathcal{H}}^i(t_1),[\hat{\mathcal{H}}^i(t_2), \ldots [\hat{\mathcal{H}}^i(t_n),\hat{\rho}^i(t_0)]\cdots]]. \tag{15}$$

The latter constitutes the starting point of the quantum Monte Carlo method for the study of charge-transport phenomena in semiconductors.[23]

In order to introduce the so-called adiabatic or Markov approximation, let us now focus on the time integral in Eq. (14). Here, the two quantities to be integrated over t' are the interaction Hamiltonian $\hat{\mathcal{H}}^i$ and the density-matrix operator $\hat{\rho}^i$. In the spirit of the perturbation approach previously recalled, the time variation of $\hat{\rho}^i$ can be considered adiabatically slow compared to that of the Hamiltonian $\hat{\mathcal{H}}^i$ within the interaction picture; indeed, the latter will exhibit rapid oscillations due to the noninteracting unitary transformation $\hat{U}_o$. As a result, the density-matrix operator $\hat{\rho}^i$ can be taken out of the time integral and evaluated at the current time t.

Within such adiabatic limit we get the following effective Liouville-von Neumann equation:

$$\frac{d}{dt}\hat{\rho}^i(t) = -i[\hat{\mathcal{H}}^i(t),\hat{\rho}^i(t_0)] - [\hat{\mathcal{H}}^i(t),[\hat{\mathcal{K}}^i(t),\hat{\rho}^i(t)]] \tag{16}$$

with

$$\hat{\mathcal{K}}^i(t) = \int_{t_0}^{t} \hat{\mathcal{H}}^i(t')dt'. \tag{17}$$

The above equation still has the double-commutator structure in Eq. (14) but it is now local in time.

Going back to the original Schrödinger picture, we finally get

$$\frac{d\hat{\rho}}{dt} = \tilde{\mathcal{L}}(\hat{\rho}) + \hat{C}, \tag{18}$$

where

$$\tilde{\mathcal{L}}(\hat{\rho}) = \frac{1}{i\hbar}[\hat{H}_o,\hat{\rho}] - [\hat{\mathcal{H}},[\hat{\mathcal{K}},\hat{\rho}]] \equiv \frac{1}{i\hbar}[\hat{H}_o,\hat{\rho}] + \Gamma(\hat{\rho}), \tag{19}$$

with

$$\hat{\mathcal{K}} = \int_{t_0}^{t} dt'\, \hat{U}_o(t-t')\hat{\mathcal{H}}\hat{U}_o^{\dagger}(t-t') \tag{20}$$

the effective Liouville superoperator within our approximation scheme, and

$$\hat{C}(t) = -i[\hat{\mathcal{H}},\hat{U}_o(t-t_0)\hat{\rho}(t_0)\hat{U}_o^{\dagger}(t-t_0)]. \tag{21}$$

The time-dependent operator $\hat{C}$ in Eq. (21) describes how the quantum-correlation effects at the initial time t_0 propagate to the current time t; indeed, combining Eqs. (6) and (21), the latter can be rewritten as

$$\hat{C}(t) = -i[\hat{\mathcal{H}},\hat{S}(t-t_0)\hat{\rho}(t)\hat{S}^{\dagger}(t-t_0)] = -i[\hat{\mathcal{H}},\hat{\rho}^i(t)], \tag{22}$$

where $\hat{S}(t-t_0)=\hat{U}_o(t-t_0)\hat{U}^{\dagger}(t-t_0)$ is the unitary transformation connecting the time evolution of the density-matrix operator in the Schrödinger and interaction pictures. This clearly shows that the initial quantum-mechanical correlations propagate from t_0 to t via the interaction-free dynamics described by the density-matrix operator written in the interaction picture. As we shall see, the above quantum-correlation operator is responsible for a number of purely quantum-mechanical phenomena, like Hartree-Fock single-particle renormalizations and coherent phonon effects.[3]

The general solution of Eq. (18) is of the form:

$$\hat{\rho}(t) = T[e^{\int_{t_0}^{t}\tilde{\mathcal{L}}(t')dt'}]\hat{\rho}(t_0) + \int_{t_0}^{t} T[e^{\int_{t'}^{t}\tilde{\mathcal{L}}(t'')dt''}]\hat{C}(t')dt', \tag{23}$$

where $T[\cdots]$ is the usual time- or chronological-ordering operator.[15,17]

At this point a few comments are in order. So far, the only approximation introduced in our theoretical description is the adiabatic decoupling between free carrier evolution and various many-body interactions; this leads to a significant modification of the system dynamics: while the exact quantum-mechanical evolution in Eq. (6) corresponds to a fully reversible and isoentropic unitary transformation, the instantaneous double-commutator structure in Eq. (19) describes, in general, a nonreversible (i.e., nonunitary) dynamics [see Eq. (23)] characterized by energy relaxation and dephasing; it follows that the system quantum entropy in Eq. (8) is no more a constant. At this level of description this behavior is totally nonphysical, clearly showing the potential failure and intrinsic limitations of the Markov approximation. However, as discussed below (see Sec. II F), the Markov limit previously introduced is usually employed together with a reduced description of the system, for which such irreversible dynamics is physically justified.

Let us finally focus on the nature of the effective Liouville superoperator in Eq. (19). As stressed before, this is the sum of a single-commutator term plus a double-commutator contribution. In the absence of carrier-carrier as well as carrier-quasiparticle interactions, i.e., $\hat{H}'=0$, the second term vanishes and the system undergoes a reversible unitary transformation induced by the single-commutator term,

which preserves the trace and the positive character of our density-matrix operator $\hat{\rho}$. In contrast, the perturbation Hamiltonian $\hat{H}'$ within the Markov limit previously introduced will induce, in general, a nonunitary evolution. Since any effective Liouville superoperator should describe correctly the time evolution of $\hat{\rho}$ and since the latter, by definition, needs to be trace-invariant and positive-definite at any time, it is important to determine if—and under which conditions—the superoperator $\tilde{\mathcal{L}}$ fulfills these two basic requirements.

As far as the first issue is concerned, recalling that the trace of a commutator is always equal to zero and taking the trace of Eq. (18), it is easy to verify that the time-derivative of the trace of $\hat{\rho}$ is equal to zero, i.e., that our effective dynamics is trace-preserving.

Let us now discuss the possible positive-definite character of $\hat{\rho}$. In general, our effective Liouville superoperator does not ensure that for any initial condition the density-matrix operator will be positive-definite at any time. Indeed, it is possible to show that the double-commutator structure in Eq. (19) can be rewritten in terms of a single-commutator structure (renormalizing the free Hamiltonian $\hat{H}_{\rm o}$) and of double commutators of the form:

$$L(\hat{\rho}) = -[\hat{\mathcal{A}},[\hat{\mathcal{A}},\hat{\rho}]]. \tag{24}$$

Each of the latter represents a particular case of the so-called Lindblad superoperators, which are known to describe completely positive (CP) maps, thus preserving the positive character of our density-matrix operator. However, our Liouville superoperator can be written in terms of the difference of the Lindblad superoperators in Eq. (24), which in general is not Lindblad-like.

Since our primary goal is the investigation of quantum-transport phenomena, we shall focus on the steady-state solution of Eq. (18). It is easy to verify that the identity operator, properly normalized, $\hat{\rho}(t) \propto \hat{\mathcal{I}}$ is the stationary solution we are looking for. A closer inspection of Eq. (23) reveals that for any positive-definite and uncorrelated initial state ($\hat{C}=0$) and for a Liouville superoperator $\hat{\mathcal{L}}$ with a nonpositive eigenvalue spectrum, in the limit $t \to \infty$ the density matrix $\hat{\rho}$ will reduce to the identity operator previously mentioned. As anticipated, this clearly shows that within such an approximation scheme our effective dynamics describes a sort of decoherence/dephasing, since possible nondiagonal terms of the density matrix will vanish on the long-time scale. This is again an artifact of the Markov limit.

Let us finally discuss the physical meaning of the steady-state solution $\hat{\rho}(t \to \infty) \propto \hat{\mathcal{I}}$. Within our noninteracting carrier-plus-quasiparticle basis λ we have $\rho_{\lambda_1\lambda_2}(t \to \infty) \propto \delta_{\lambda_1\lambda_2}$. This tells us that, physically speaking, the steady-state solution of our transport equation corresponds to an equally probable population of all the microscopic states λ without any interstate quantum coherence ($\rho_{\lambda_1 \neq \lambda_2}=0$). This scenario is typical of the present global (carrier+quasiparticle) description; in contrast, within a reduced description (of the carrier subsystem only) the steady-state solution differs from the identity operator, since in this case the trace over nonrelevant degrees of freedom will translate into a thermal weight over our electronic states (see Sec. II F).

D. Generalized scattering superoperator

Let us now evaluate the explicit form of the scattering superoperator Γ in Eq. (20). The effective Liouville-von Neumann equation in Eq. (18) can be easily rewritten in the noninteracting-states basis $\{|\lambda\rangle\}$ previously introduced:

$$\frac{d\rho_{\lambda_1\lambda_2}}{dt} = \frac{\epsilon_{\lambda_1} - \epsilon_{\lambda_2}}{i\hbar}\rho_{\lambda_1\lambda_2} + C_{\lambda_1\lambda_2} + \sum_{\lambda_1'\lambda_2'} \Gamma_{\lambda_1\lambda_2,\lambda_1'\lambda_2'}\rho_{\lambda_1'\lambda_2'}. \tag{25}$$

In order to derive the explicit form of the superoperator matrix elements $\Gamma_{\lambda_1\lambda_2,\lambda_1'\lambda_2'}$, let us expand the double commutator in Eq. (20):

$$\begin{aligned}\sum_{\lambda_1'\lambda_2'} \Gamma_{\lambda_1\lambda_2,\lambda_1'\lambda_2'}\rho_{\lambda_1'\lambda_2'} &= [\hat{\mathcal{H}},[\hat{\mathcal{K}},\hat{\rho}]]_{\lambda_1\lambda_2} \\ &= \sum_{\lambda_1'\lambda_2'} (\mathcal{H}_{\lambda_1\lambda_1'}\rho_{\lambda_1'\lambda_2'}\mathcal{K}_{\lambda_2'\lambda_2} \\ &\quad + \mathcal{K}_{\lambda_1\lambda_1'}\rho_{\lambda_1'\lambda_2'}\mathcal{H}_{\lambda_2'\lambda_2}) \\ &\quad - \sum_{\lambda_1'\lambda_2'} (\mathcal{H}_{\lambda_1\lambda_1'}\mathcal{K}_{\lambda_1'\lambda_2'}\rho_{\lambda_2'\lambda_2} \\ &\quad + \rho_{\lambda_1\lambda_1'}\mathcal{K}_{\lambda_1'\lambda_2'}\mathcal{H}_{\lambda_2'\lambda_2}).\end{aligned} \tag{26}$$

As we can see, the scattering operator Γ can be written as the difference of the following "in-" and "out-scattering" terms:

$$\Gamma^{\rm in}_{\lambda_1\lambda_2,\lambda_1'\lambda_2'} = \mathcal{H}_{\lambda_1\lambda_1'}\mathcal{K}_{\lambda_2'\lambda_2} + \mathcal{K}_{\lambda_1\lambda_1'}\mathcal{H}_{\lambda_2'\lambda_2}, \tag{27}$$

$$\Gamma^{\rm out}_{\lambda_1\lambda_2,\lambda_1'\lambda_2'} = \sum_{\lambda''} \mathcal{H}_{\lambda_1\lambda''}\mathcal{K}_{\lambda''\lambda_1'}\delta_{\lambda_2\lambda_2'} + \sum_{\lambda''} \delta_{\lambda_1\lambda_1'}\mathcal{K}_{\lambda_2'\lambda''}\mathcal{H}_{\lambda''\lambda_2}. \tag{28}$$

For the case of a time-independent perturbation $\hat{H}' = \hbar\hat{\mathcal{H}}$, the operator $\hat{\mathcal{K}}$ can be rewritten as

$$\hat{\mathcal{K}} = \int_0^{t-t_0} d\tau \hat{U}_{\rm o}(\tau)\hat{\mathcal{H}}\hat{U}_{\rm o}^{\dagger}(\tau). \tag{29}$$

Taking into account that within the λ representation the noninteracting evolution operator $\hat{U}_{\rm o}$ is simply given by

$$U_{\rm o}^{\lambda\lambda'}(\tau) = e^{\epsilon_\lambda \tau/i\hbar}\delta_{\lambda\lambda'}, \tag{30}$$

the matrix elements of the operator $\hat{\mathcal{K}}$ in Eq. (29) result to be

$$\mathcal{K}_{\lambda\lambda'} = 2\pi\mathcal{H}_{\lambda\lambda'}\mathcal{D}_{\lambda\lambda'} \tag{31}$$

with

$$\mathcal{D}_{\lambda\lambda'} = \frac{1}{2\pi}\int_0^{t-t_0} e^{(\epsilon_\lambda-\epsilon_{\lambda'})\tau/i\hbar}d\tau = \mathcal{D}^*_{\lambda'\lambda}. \tag{32}$$

By inserting the above result into Eqs. (27) and (28) and recalling that $\mathcal{H}_{\lambda\lambda'} \equiv H'_{\lambda\lambda'}/\hbar = \mathcal{H}^*_{\lambda'\lambda}$, we finally obtain:

$$\Gamma^{\text{in}}_{\lambda_1\lambda_2,\lambda_1'\lambda_2'} = \frac{2\pi}{\hbar^2}(H'_{\lambda_1\lambda_1'}H'^*_{\lambda_2\lambda_2'}\mathcal{D}^*_{\lambda_2\lambda_2'} + \mathcal{D}_{\lambda_1\lambda_1'}H'_{\lambda_1\lambda_1'}H'^*_{\lambda_2\lambda_2'}); \tag{33}$$

$$\Gamma^{\text{out}}_{\lambda_1\lambda_2,\lambda_1'\lambda_2'} = \frac{2\pi}{\hbar^2} \times\sum_{\lambda''} (H'^*_{\lambda''\lambda_1}H'_{\lambda''\lambda_1'}\mathcal{D}_{\lambda''\lambda_1'}\delta_{\lambda_2\lambda_2'} + \delta_{\lambda_1\lambda_1'}\mathcal{D}^*_{\lambda''\lambda_2'}H'^*_{\lambda''\lambda_2'}H'_{\lambda''\lambda_2}). \tag{34}$$

In general, the scattering superoperator Γ is a function of time; however, in the limit $t_0 \to -\infty$—also called "completed-collision limit"[20,23]—the function $\mathcal{D}$ in Eq. (32) becomes time-independent:

$$\mathcal{D}^{-\infty}_{\lambda\lambda'} = \frac{1}{2\pi}\int_0^{\infty} e^{(\epsilon_\lambda-\epsilon_{\lambda'})\tau/i\hbar}d\tau. \tag{35}$$

It follows that in this limit the operator $\mathcal{K}$ as well as the superoperators Γ and $\tilde{\mathcal{L}}$ are also time-independent. In this case there is no need for the time-ordering operator T in Eq. (23). Moreover, the real part of the function $\mathcal{D}^{-\infty}$ in Eq. (35) gives the well-known energy-conserving Dirac delta function, i.e.,

$$\mathcal{D}^{-\infty}_{\lambda\lambda'} = \frac{\hbar}{2}\delta(\epsilon_\lambda - \epsilon_{\lambda'}) + \mathcal{R}_{\lambda\lambda'}, \tag{36}$$

while the imaginary part—denoted by $\mathcal{R}_{\lambda\lambda'}$—describes, in general, energy-renormalization effects. Within the validity limits of the present Markov treatment, such renormalization effects can be safely neglected: if, as requested, the perturbation Hamiltonian is small compared to the noninteracting one, then the resulting energy-level renormalization is small compared to the noninteracting energy levels ϵ_λ.

At this point a few comments on the evaluation of the time integral in Eq. (35) are in order. Indeed, it is well-known that the limit $t_0 \to -\infty$ needs to be performed properly; more specifically, this is realized by adding to the energy difference an infinitesimally small imaginary part, which ensures the convergence of the time integration. A qualitative—but not rigorous—interpretation of such a mathematical prescription is based on the so-called "adiabatic switching-on" procedure:[26] The idea is that, starting from $t=-\infty$, the interaction mechanism/Hamiltonian is slowly or adiabatically switched on. By employing the nonequilibrium Green's function formalism, it is possible to show[17] that such an imaginary part is not an artificial ingredient: it corresponds to the imaginary part of the electron self-energy, thus describing the finite lifetime of our electronic states due to all relevant interaction mechanisms. A proper account of such an effect—not relevant in the present discussion—leads to apparent violations of the energy-conserving transitions predicted by the Dirac delta function in Eq. (36), the so-called "collisional broadening."

As previously recalled, it is imperative to establish if—and under which conditions—the scattering superoperator in Eq. (26) preserves the positive-definite nature of our density-matrix operator ρ. As discussed in the Appendix:

(i) contrary to the semiclassical or Boltzmann dynamics (see Sec. II E), the effective Liouville superoperator previously identified does not correspond to a so-called "CP map," i.e., it does not preserve, in general, the positive-definite character of our density-matrix operator;

(ii) its eigenvalue spectrum, i.e.,

$$\tilde{\mathcal{L}}(\hat{\rho}) = \Lambda\hat{\rho}, \tag{37}$$

always contains the $\Lambda=0$ eigenvalue, which corresponds to the steady-state transport solution; and

(iii) in the "small-coupling limit" it is possible to show that the steady-state density-matrix operator—corresponding to the $\Lambda=0$ eigenvalue—is always positive definite, i.e.,

$$\hat{\rho}^{\Lambda=0} = \sum_{\bar{\lambda}} P_{\bar{\beta}}|\bar{\beta}\rangle\langle\bar{\beta}|, \tag{38}$$

where the basis states $|\bar{\beta}\rangle$ are the eigenvectors of $\hat{\rho}^{\Lambda=0}$, and $P_{\bar{\beta}}$ is a (non-negative) probability distribution.

E. Semiclassical limit

The well-known semiclassical or Boltzmann transport theory[1] can be easily derived from the quantum-transport formulation presented so far by introducing the so-called diagonal or semiclassical limit. The latter corresponds to neglecting all nondiagonal density-matrix elements (and therefore any quantum-mechanical phase coherence between the generic states λ_1 and λ_2), i.e.,

$$\rho_{\lambda_1\lambda_2} = f_{\lambda_1}\delta_{\lambda_1\lambda_2}, \tag{39}$$

where the diagonal elements f_λ describe the semiclassical distribution function over the noninteracting basis states λ.

By introducing the above semiclassical density matrix into Eq. (25) for the diagonal elements ($\lambda=\lambda_1=\lambda_2$), and inserting the explicit form of the elements $\Gamma_{\lambda\lambda}$ of the scattering operator in the limit $t_0 \to -\infty$ [see Eq. (35)], we finally obtain the usual form of the Boltzmann transport equation written in our basis states:

$$\frac{df_\lambda}{dt} = \sum_{\lambda'} (P_{\lambda\lambda'}f_{\lambda'} - P_{\lambda'\lambda}f_\lambda), \tag{40}$$

where

$$P_{\lambda\lambda'} = P_{\lambda'\lambda} = \frac{2\pi}{\hbar}|H'_{\lambda'\lambda}|^2\delta(\epsilon_{\lambda'} - \epsilon_\lambda) \tag{41}$$

are the semiclassical scattering rates given by the well-known Fermi's golden rule. In addition to the square of the interaction matrix element $H'_{\lambda'\lambda}$, they contain the energy-conserving Dirac delta function:

$$\delta(\epsilon_{\lambda'} - \epsilon_\lambda) = \frac{\mathcal{D}^{-\infty}_{\lambda\lambda'} + \mathcal{D}^{-\infty}_{\lambda'\lambda}}{\hbar} = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i(\epsilon_{\lambda'}-\epsilon_\lambda)\tau} d\tau. \quad (42)$$

Within the semiclassical limit the free-rotation term in Eq. (25) vanishes, and the same applies to the quantum-correlation contributions $C_{\lambda\lambda}$.

Our analysis shows that the quantum-transport equation in Eq. (25) can be regarded as the quantum-mechanical generalization of the Boltzmann equation in Eq. (40). Indeed, the in- and out-scattering superoperators in Eq. (26) are the quantum-mechanical generalizations of the standard in- and out-scattering terms entering the Boltzmann collision operator in Eq. (40).

As a confirmation of the fact that the Markov approximation leads to a totally nonphysical nonreversible (i.e., nonunitary) system evolution, it is possible to show that the system entropy S in Eq. (8) is a nondecreasing function of time.

We stress that, contrary to the usual semiclassical transport theory, the Boltzmann-like equation in Eq. (40) describes a scattering dynamics within the whole $\lambda=\{n_\alpha\},\{n_{\mathbf{q}}\}$ space, i.e., the generic scattering probability $P_{\lambda\lambda'}$ in Eq. (41) describes a transition from the state $\{n_\alpha\},\{n_{\mathbf{q}}\}$ to the state $\{n_{\alpha'}\},\{n_{\mathbf{q}'}\}$. In other words, so far no reduction procedure to the α-subsystem has been performed; it follows that for a given transition of the α subsystem ($\{n_\alpha\}\to\{n_{\alpha'}\}$), a corresponding transition ($\{n_{\mathbf{q}}\}\to\{n_{\mathbf{q}'}\}$) of the quasiparticle subsystem will also take place. This explains why, contrary to the usual Boltzmann theory, the scattering probabilities in Eq. (41) are symmetric, i.e., invariant under time reversal: $P_{\lambda\lambda'}=P_{\lambda'\lambda}$; moreover, the Dirac delta function in Eq. (41) leads to the conservation of the total energy of the system.

A second important remark is that, contrary to the nondiagonal density-matrix description previously introduced, the Markov limit combined with the semiclassical or diagonal approximation in Eq. (39) ensures that at any time t our semiclassical distribution function f_λ is always positive-definite (see the Appendix).

Let us finally discuss the steady-state solution of the Boltzmann transport equation in Eq. (40). From the detailed-balance principle, i.e.,

$$P_{\lambda\lambda'} f_{\lambda'} = P_{\lambda'\lambda} f_\lambda, \quad (43)$$

and considering that the semiclassical scattering rates in Eq. (41) are symmetric ($P_{\lambda\lambda'}=P_{\lambda'\lambda}$), we get

$$\frac{f_{\lambda'}}{f_\lambda} = \frac{P_{\lambda\lambda'}}{P_{\lambda'\lambda}} = 1. \quad (44)$$

Exactly as in the quantum-mechanical case, the steady-state solution corresponds to a uniform distribution over the noninteracting carrier-plus-quasiparticle states: $f_\lambda \propto \delta_{\lambda\lambda}$. As discussed in Sec. II F, this is not the case when our kinetic description is reduced to the α subsystem only.

F. Reduced description

As discussed in Sec. II B, the average value of any given physical quantity A can be easily expressed in terms of the density-matrix operator $\hat{\rho}$ according to Eq. (3). In the study of charge-transport phenomena in semiconductor nanostructures, most of the physical quantities of interest depend on the electronic-subsystem coordinates α only (carrier drift velocity, total electronic energy, carrier-carrier correlation function, etc.), i.e.,

$$A_{\{n_\alpha\},\{n_{\mathbf{q}}\};\{n_{\alpha'}\},\{n_{\mathbf{q}'}\}} = A_{\{n_\alpha\},\{n_{\alpha'}\}}\delta_{\{n_{\mathbf{q}}\},\{n_{\mathbf{q}'}\}}. \quad (45)$$

In this case it is convenient to write

$$A = \sum_{\lambda\lambda'} A_{\lambda\lambda'}\rho_{\lambda'\lambda} = \sum_{\{n_\alpha\},\{n_{\alpha'}\}} A_{\{n_\alpha\},\{n_{\alpha'}\}}\rho^c_{\{n_{\alpha'}\},\{n_\alpha\}}, \quad (46)$$

where

$$\rho^c_{\{n_\alpha\},\{n_{\alpha'}\}} = \sum_{\{n_{\mathbf{q}}\}} \rho_{\{n_\alpha\},\{n_{\mathbf{q}}\};\{n_{\alpha'}\},\{n_{\mathbf{q}}\}} \quad (47)$$

is the so-called reduced or electronic density matrix. Equation (47) can also be written in an operatorial form as

$$\hat{\rho}^c = \mathrm{tr}\{\hat{\rho}\}_{\{n_{\mathbf{q}}\}}, \quad (48)$$

which shows that the electronic density-matrix operator $\hat{\rho}^c$ is obtained by performing a trace operation over the quasiparticle coordinates $\mathbf{q}$. Since $\hat{\rho}^c$ is the only quantity entering the evaluation of the average value in Eq. (46), it is desirable to derive a corresponding equation of motion for the reduced density-matrix operator. Combining Eqs. (18) and (48) we get

$$\frac{d\hat{\rho}^c}{dt} = \mathrm{tr}\{\tilde{\mathcal{L}}(\hat{\rho})\}_{\{n_{\mathbf{q}}\}} + \mathrm{tr}\{\hat{C}\}_{\{n_{\mathbf{q}}\}}. \quad (49)$$

In general, the trace over the quasiparticle coordinates does not commute with the Liouville superoperator $\tilde{\mathcal{L}}$ in Eq. (19), which does not allow one to obtain a closed equation of motion for the reduced density-matrix operator $\hat{\rho}^c$. This clearly does not apply when the interaction Hamiltonian is a function of the carrier coordinates only, e.g., carrier-carrier, carrier-impurity, etc. In contrast, in the presence of carrier-quasiparticle coupling additional approximations are needed. In order to get a closed equation of motion for the reduced density-matrix operator, the typical assumption is to consider the quasiparticle subsystem as characterized by a huge number of degrees of freedom (compared to the subsystem α). In other words this amounts to say that the $\mathbf{q}$ subsystem has an infinitely high heat capacity, i.e., it behaves as a thermal bath; this allows one to consider the quasiparticle subsystem always in thermal equilibrium, i.e., not significantly perturbed by the carrier subsystem α. Within such approximation scheme, the global ($\alpha+\mathbf{q}$) density-matrix operator $\hat{\rho}$ can be written as the product of the equilibrium density-matrix operator for the quasiparticle subsystem $\hat{\rho}^{qp}$ and the reduced density-matrix operator $\hat{\rho}^c$:

$$\hat{\rho} = \hat{\rho}^c \otimes \hat{\rho}^{qp}, \quad \hat{\rho}^{qp} = \frac{e^{-\hat{H}^{qp}_{\mathrm{o}}/k_{\mathrm{B}}T}}{\mathrm{tr}\left\{e^{-\hat{H}^{qp}_{\mathrm{o}}/k_{\mathrm{B}}T}\right\}}. \quad (50)$$

The corresponding matrix elements within our basis states $\lambda=\{n_\alpha\},\{n_{\mathbf{q}}\}$ are then given by

$$\rho_{\lambda\lambda'} = \rho^c_{\{n_\alpha\}\{n_{\alpha'}\}} f^{qp}_{\{n_{\mathbf{q}}\}} \delta_{\{n_{\mathbf{q}}\}\{n_{\mathbf{q}'}\}} \tag{51}$$

with

$$f^{qp}_{\{n_{\mathbf{q}}\}} = \frac{e^{-\epsilon_{\{n_{\mathbf{q}}\}}/k_B T}}{\sum_{\{n_{\mathbf{q}}\}} e^{-\epsilon_{\{n_{\mathbf{q}}\}}/k_B T}}. \tag{52}$$

By inserting these density-matrix elements into Eq. (25) and performing the trace over the quasiparticle coordinates, it is easy to get the following effective Liouville-von Neumann equation for the reduced density matrix ρ^c:

$$\frac{d\rho^c_{\{n_{\alpha_1}\}\{n_{\alpha_2}\}}}{dt} = \frac{\epsilon_{\{n_{\alpha_1}\}} - \epsilon_{\{n_{\alpha_2}\}}}{i\hbar} \rho^c_{\{n_{\alpha_1}\}\{n_{\alpha_2}\}} + C^c_{\{n_{\alpha_1}\}\{n_{\alpha_2}\}} + \sum_{\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}} \Gamma^c_{\{n_{\alpha_1}\}\{n_{\alpha_2}\},\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}} \rho^c_{\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}} \tag{53}$$

with

$$\Gamma^c_{\{n_{\alpha_1}\}\{n_{\alpha_2}\},\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}} = \sum_{\{n_{\mathbf{q}}\}\{n_{\mathbf{q}'}\}} \times \Gamma_{\{n_{\alpha_1}\}\{n_{\mathbf{q}}\},\{n_{\alpha_2}\}\{n_{\mathbf{q}}\};\{n_{\alpha_1'}\}\{n_{\mathbf{q}'}\},\{n_{\alpha_2'}\}\{n_{\mathbf{q}'}\}} f^{qp}_{\{n_{\mathbf{q}'}\}} \tag{54}$$

and

$$C^c_{\{n_{\alpha_1}\}\{n_{\alpha_2}\}} = \sum_{\{n_{\mathbf{q}}\}} C_{\{n_{\alpha_1}\}\{n_{\mathbf{q}}\},\{n_{\alpha_2}\}\{n_{\mathbf{q}}\}}. \tag{55}$$

By denoting with Γ^c the effective scattering superoperator defined by the matrix elements in Eq. (54)—acting on the α Hilbert subspace only—the new effective Liouville superoperator (i.e., traced over the $\mathbf{q}$ coordinates) is given by

$$\tilde{\mathcal{L}}^c(\hat{\rho}^c) = \frac{1}{i\hbar}[\hat{H}^c_o, \hat{\rho}^c] + \Gamma^c(\hat{\rho}^c). \tag{56}$$

The latter, however, does not contain the double-commutator structure previously discussed; this aspect will be more extensively addressed in the following.

For all relevant carrier-quasiparticle interaction mechanisms in semiconductor nanostructures—e.g., carrier-phonon, carrier-plasmon, etc.—the perturbation Hamiltonian $\hat{H}'$ can be written as

$$\hat{H}' = \hbar\hat{\mathcal{H}} = \hbar\sum_{\mathbf{q}} (\hat{\mathcal{H}}_{\mathbf{q}}\hat{b}_{\mathbf{q}} + \hat{\mathcal{H}}^\dagger_{\mathbf{q}}\hat{b}^\dagger_{\mathbf{q}}). \tag{57}$$

Here $\hat{\mathcal{H}}_{\mathbf{q}} = \hat{\mathcal{H}}^\dagger_{-\mathbf{q}}$ are electronic operators (parameterized by the quasiparticle wave vector $\mathbf{q}$) acting on the α subsystem only. The two terms in Eq. (57)—corresponding to quasiparticle destruction and creation—describe quasiparticle absorption and emission processes.

Let us consider again the definition of the effective carrier-quasiparticle scattering superoperator Γ^c in Eq. (54) written in operatorial form, i.e.,

$$\Gamma^c(\hat{\rho}^c) = -\mathrm{tr}\{[\hat{\mathcal{H}},[\hat{\mathcal{K}},\hat{\rho}^c\hat{\rho}^{qp}]]\}_{\{n_{\mathbf{q}}\}}, \tag{58}$$

where the double-commutator form in Eq. (20) has been introduced.

By inserting into Eqs. (29) and (58) the explicit form of the carrier-quasiparticle Hamiltonian in Eq. (57) and using the bosonic commutation relations $[\hat{b}_{\mathbf{q}}, \hat{b}^\dagger_{\mathbf{q}'}] = \delta_{\mathbf{q}\mathbf{q}'}$, we obtain an explicit form of the effective carrier-quasiparticle scattering superoperator in Eq. (58). More specifically we get

$$\Gamma^c(\hat{\rho}^c) = -\sum_{\mathbf{q}\pm} \left(N_{\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2}\right)[\hat{\mathcal{H}}_{\mathbf{q}}, \hat{\mathcal{K}}^\pm_{\mathbf{q}}\hat{\rho}^c - \hat{\rho}^c\hat{\mathcal{K}}^\mp_{\mathbf{q}}] \tag{59}$$

with

$$\hat{\mathcal{K}}^\pm_{\mathbf{q}} = \int_0^{t-t_0} d\tau e^{\hat{H}^c_o \tau/i\hbar} \hat{\mathcal{H}}^\dagger_{\mathbf{q}} e^{-\hat{H}^c_o \tau/i\hbar} e^{\pm\epsilon_{\mathbf{q}}\tau/i\hbar}. \tag{60}$$

Here

$$N_{\mathbf{q}} = \mathrm{tr}\{\hat{b}^\dagger_{\mathbf{q}}\hat{b}_{\mathbf{q}}\hat{\rho}^{qp}\} = \frac{1}{e^{\epsilon_{\mathbf{q}}/k_B T} - 1} \tag{61}$$

denotes the equilibrium average occupation number for the quasiparticle state $\mathbf{q}$. As we can see, for each quasiparticle state $\mathbf{q}$ we have two contributions ($\pm$) describing quasiparticle emission and absorption.

The effective scattering superoperator Γ^c in Eq. (59) does not exhibit the double-commutator structure typical of the global description [see Eq. (20)]. Indeed, denoting with

$$\hat{\mathcal{K}}^\pm_1 = \frac{1}{2}(\hat{\mathcal{K}}^\pm_{\mathbf{q}} + \hat{\mathcal{K}}^\mp_{\mathbf{q}}), \quad \hat{\mathcal{K}}^\pm_2 = \frac{1}{2}(\hat{\mathcal{K}}^\pm_{\mathbf{q}} - \hat{\mathcal{K}}^\mp_{\mathbf{q}}), \tag{62}$$

the superoperator Γ^c in Eq. (59) can also be expressed as

$$\Gamma^c(\hat{\rho}^c) = -\sum_{\mathbf{q}\pm} \left(N_{\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2}\right)([\hat{\mathcal{H}}_{\mathbf{q}}, [\hat{\mathcal{K}}^\pm_1, \hat{\rho}^c]] + [\hat{\mathcal{H}}_{\mathbf{q}}, \{\hat{\mathcal{K}}^\pm_2, \hat{\rho}^c\}]). \tag{63}$$

As anticipated, the scattering superoperator involves again a double-commutator term, but we have also a commutator-anticommutator contribution.

To better underline the physical role played by the above double-commutator versus commutator-anticommutator contributions, let us recall a simplified model usually invoked to qualitatively describe the quantum-mechanical evo lution of open systems, i.e., subsystems interacting with their environment. Within the Schrödinger picture, the latter are typically treated by adding to the system Hamiltonian $\hat{H}$ an anti-Hermitian part (imaginary potential) $\hat{V}^{\mathrm{env}}$ describing the system-environment coupling:

$$i\hbar\frac{d}{dt}|\psi\rangle = (\hat{H} + \hat{V}^{\text{env}})|\psi\rangle. \tag{64}$$

Starting from the above modified Schrödinger equation, it is easy to obtain a corresponding version of the Liouville-von Neumann equation in Eq. (5):

$$\frac{d\hat{\rho}}{dt} = \frac{1}{i\hbar}[\hat{H},\hat{\rho}] + \frac{1}{i\hbar}\{\hat{V}^{\text{env}},\hat{\rho}\}. \tag{65}$$

In addition to the commutatorlike dynamics typical of a closed system, we deal with an anticommutator term, describing dissipation induced by the system-environment coupling. Indeed, contrary to the closed dynamics in Eq. (5), the latter leads to a nonreversible dynamics. Such a simplified model is known to be highly nonphysical, since it does not preserve the trace of the density-matrix operator; however, it clearly shows how the commutator structure is intimately related to a closed evolution, while anticommutator terms always describe dissipation processes.

Contrary to the above simplified model, the scattering superoperator in Eq. (59)—in view of its outercommutator structure—is trace-preserving. However, the presence of the commutator-anticommutator contribution is a clear fingerprint of carrier-quasiparticle dissipation phenomena leading to genuine energy-relaxation/dephasing processes.

As far as the correlation term in Eq. (55) is concerned, it is easy to verify that the latter vanishes for the linear-coupling carrier-quasiparticle Hamiltonian in Eq. (57).

In summary, within the approximation scheme considered so far we get the following effective equation of motion for the reduced density-matrix operator $\hat{\rho}^c$:

$$\frac{d\hat{\rho}^c}{dt} = \tilde{\mathcal{L}}^c(\hat{\rho}^c) \tag{66}$$

with $\hat{\mathcal{L}}^c$ defined in Eq. (56). In the limit $t_0 \to -\infty$, the effective Liouville superoperator in Eq. (56) becomes time-independent, and the general solution of the homogeneous equation in Eq. (66) is of the form

$$\hat{\rho}^c(t) = e^{\tilde{\mathcal{L}}^c(t-t_0)}\hat{\rho}^c(t_0). \tag{67}$$

Again, contrary to the isoentropic and fully reversible unitary evolution in Eq. (6), the instantaneous double-commutator plus commutator-anticommutator structures in Eq. (63) describe a nonreversible (i.e., nonunitary) dynamics characterized by energy relaxation and dephasing induced by the carrier-quasiparticle coupling in Eq. (57).

We shall now derive the explicit form of the scattering superoperator Γ^c within our noninteracting-carrier basis $\{n_\alpha\}$. To this end let us expand the various terms entering Eq. (59):

$$\begin{aligned}\sum_{\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}} \Gamma^c_{\{n_{\alpha_1}\}\{n_{\alpha_2}\},\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}}\rho^c_{\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}} &= \sum_{\mathbf{q}\pm}\left(N_{\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2}\right)\\ &\times \sum_{\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}} [(\mathcal{H}^{\mathbf{q}}_{\{n_{\alpha_1}\}\{n_{\alpha_1'}\}}\rho^c_{\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}}\mathcal{K}^{\mathbf{q}\mp}_{\{n_{\alpha_2'}\}\{n_{\alpha_2}\}}\\ &+ \mathcal{K}^{\mathbf{q}\pm}_{\{n_{\alpha_1}\}\{n_{\alpha_1'}\}}\rho^c_{\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}}\mathcal{H}^{\mathbf{q}}_{\{n_{\alpha_2'}\}\{n_{\alpha_2}\}})\\ &- (\mathcal{H}^{\mathbf{q}}_{\{n_{\alpha_1}\}\{n_{\alpha_1'}\}}\mathcal{K}^{\mathbf{q}\pm}_{\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}}\rho^c_{\{n_{\alpha_2'}\}\{n_{\alpha_2}\}}\\ &+ \rho^c_{\{n_{\alpha_1}\}\{n_{\alpha_1'}\}}\mathcal{K}^{\mathbf{q}\mp}_{\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}}\mathcal{H}^{\mathbf{q}}_{\{n_{\alpha_2'}\}\{n_{\alpha_2}\}})].\end{aligned} \tag{68}$$

As we can see, also in this case the scattering superoperator Γ^c can be regarded as the difference of the following in-and out-scattering terms:

$$\begin{aligned}\Gamma^{\text{in}}_{\{n_{\alpha_1}\}\{n_{\alpha_2}\},\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}} &= \sum_{\mathbf{q}\pm}\left(N_{\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2}\right)\\ &\times(\mathcal{H}^{\mathbf{q}}_{\{n_{\alpha_1}\}\{n_{\alpha_1'}\}}\mathcal{K}^{\mathbf{q}\mp}_{\{n_{\alpha_2'}\}\{n_{\alpha_2}\}}\\ &+ \mathcal{K}^{\mathbf{q}\pm}_{\{n_{\alpha_1}\}\{n_{\alpha_1'}\}}\mathcal{H}^{\mathbf{q}}_{\{n_{\alpha_2'}\}\{n_{\alpha_2}\}}),\end{aligned} \tag{69}$$

$$\begin{aligned}\Gamma^{\text{out}}_{\{n_{\alpha_1}\}\{n_{\alpha_2}\},\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}} &= \sum_{\{n_{\alpha''}\},\mathbf{q}\pm}\left(N_{\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2}\right)\\ &\times(\mathcal{H}^{\mathbf{q}}_{\{n_{\alpha_1}\}\{n_{\alpha''}\}}\mathcal{K}^{\mathbf{q}\pm}_{\{n_{\alpha''}\}\{n_{\alpha_1'}\}}\delta_{\{n_{\alpha_2}\}\{n_{\alpha_2'}\}}\\ &+ \delta_{\{n_{\alpha_1}\}\{n_{\alpha_1'}\}}\mathcal{K}^{\mathbf{q}\mp}_{\{n_{\alpha_2'}\}\{n_{\alpha''}\}}\mathcal{H}^{\mathbf{q}}_{\{n_{\alpha''}\}\{n_{\alpha_2}\}}).\end{aligned} \tag{70}$$

As for the case of the global (carrier+quasiparticle) description presented in Sec. II D, the matrix elements $\mathcal{K}^{\mathbf{q}\pm}_{\{n_\alpha\}\{n_{\alpha'}\}}$ of the operator $\hat{\mathcal{K}}^{\pm}_{\mathbf{q}}$ in Eq. (60) may be expressed in terms of the matrix elements of the operator $\hat{\mathcal{H}}_{\mathbf{q}}$:

$$\mathcal{K}^{\mathbf{q}\pm}_{\{n_\alpha\}\{n_{\alpha'}\}} = 2\pi\mathcal{H}^{\mathbf{q}*}_{\{n_{\alpha'}\}\{n_\alpha\}}\mathcal{D}^{\mathbf{q}\mp*}_{\{n_{\alpha'}\}\{n_\alpha\}} \tag{71}$$

with

$$\mathcal{D}^{\mathbf{q}\pm}_{\{n_\alpha\}\{n_{\alpha'}\}} = \frac{1}{2\pi}\int_0^{t-t_0} e^{(\epsilon_{\{n_\alpha\}}-\epsilon_{\{n_{\alpha'}\}}\pm\epsilon_{\mathbf{q}})\tau/i\hbar}d\tau. \tag{72}$$

Again, in the completed-collision limit ($t_0 \to -\infty$), the real part of $\mathcal{D}$ provides the energy-conserving Dirac delta function, while its imaginary part describes carrier-quasiparticle energy-renormalization effects. By inserting this relation into the above in- and out-scattering rates and recalling that $\mathcal{H}^{\mathbf{q}*}_{\{n_\alpha\}\{n_{\alpha'}\}} = \mathcal{H}^{-\mathbf{q}}_{\{n_{\alpha'}\}\{n_\alpha\}}$ and $\mathcal{D}^{\mathbf{q}\pm*}_{\{n_\alpha\}\{n_{\alpha'}\}} = \mathcal{D}^{\mathbf{q}\mp}_{\{n_{\alpha'}\}\{n_\alpha\}}$, we finally get

$$\begin{aligned}\Gamma^{\text{in}}_{\{n_{\alpha_1}\}\{n_{\alpha_2}\},\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}} &= \frac{2\pi}{\hbar^2}\sum_{\mathbf{q}\pm}\left(N_{\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2}\right)\\ &\times(H^{\mathbf{q}}_{\{n_{\alpha_1}\}\{n_{\alpha_1'}\}}H^{\mathbf{q}*}_{\{n_{\alpha_2}\}\{n_{\alpha_2'}\}}\mathcal{D}^{\mathbf{q}\pm*}_{\{n_{\alpha_2}\}\{n_{\alpha_2'}\}}\\ &+ \mathcal{D}^{\mathbf{q}\pm}_{\{n_{\alpha_1}\}\{n_{\alpha_1'}\}}H^{\mathbf{q}}_{\{n_{\alpha_1}\}\{n_{\alpha_1'}\}}H^{\mathbf{q}*}_{\{n_{\alpha_2}\}\{n_{\alpha_2'}\}})\end{aligned} \tag{73}$$

and

$$\begin{aligned}\Gamma^{\text{out}}_{\{n_{\alpha_1}\}\{n_{\alpha_2}\},\{n_{\alpha_1'}\}\{n_{\alpha_2'}\}} &= \frac{2\pi}{\hbar^2}\sum_{\{n_{\alpha''}\},\mathbf{q}\pm}\left(N_{\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2}\right)\\ &\times(H^{\mathbf{q}*}_{\{n_{\alpha''}\}\{n_{\alpha_1}\}}H^{\mathbf{q}}_{\{n_{\alpha''}\}\{n_{\alpha_1'}\}}\mathcal{D}^{\mathbf{q}\pm}_{\{n_{\alpha''}\}\{n_{\alpha_1'}\}}\delta_{\{n_{\alpha_2}\}\{n_{\alpha_2'}\}}\\ &+ \delta_{\{n_{\alpha_1}\}\{n_{\alpha_1'}\}}\mathcal{D}^{\mathbf{q}\pm*}_{\{n_{\alpha''}\}\{n_{\alpha_2'}\}}H^{\mathbf{q}*}_{\{n_{\alpha''}\}\{n_{\alpha_2'}\}}H^{\mathbf{q}}_{\{n_{\alpha''}\}\{n_{\alpha_2}\}}),\end{aligned} \tag{74}$$

where $H^{\mathbf{q}}_{\{n_\alpha\}\{n_{\alpha'}\}}=\hbar\mathcal{H}^{\mathbf{q}}_{\{n_\alpha\}\{n_{\alpha'}\}}$. We stress that the above in- and out-scattering superoperators are linear, i.e., ρ-independent. As we shall see, this feature—typical of the present many-electron description $\{n_\alpha\}$—will be lost in the single-particle picture discussed below (see Sec. II G).

Let us finally focus on the steady-state solution of the quantum-transport equation in Eq. (66). Contrary to the global (carrier+quasiparticle) equation in Eq. (18), the identity operator $\hat{\mathcal{I}}$ is no more a solution. Indeed, the latter fulfills the double commutator but not the commutator-anticommutator structure in Eq. (63). Moreover, as we shall see in Sec. III, the steady-state solution of our effective transport equation is in general nondiagonal. We also stress that, as for the global (carrier+quasiparticle) description, in the small-coupling limit it is possible to show that the steady-state reduced density matrix is again positive-definite (see the Appendix).

Also for the present reduced description we can consider the semiclassical or Boltzmann limit; as described in Sec. II E, this corresponds to neglecting the nondiagonal matrix elements of the reduced density matrix, i.e., $\rho^c_{\{n_{\alpha_1}\}\{n_{\alpha_2}\}}=f^c_{\{n_{\alpha_1}\}}\delta_{\{n_{\alpha_1}\}\{n_{\alpha_2}\}}$. Within such approximation scheme the quantum-transport equation in Eq. (59) reduces to the following Boltzmann equation for the carrier subsystem:

$$\frac{df^c_{\{n_\alpha\}}}{dt}=\sum_{\{n_{\alpha'}\}}(P^c_{\{n_\alpha\}\{n_{\alpha'}\}}f^c_{\{n_{\alpha'}\}}-P^c_{\{n_{\alpha'}\}\{n_\alpha\}}f^c_{\{n_\alpha\}}), \tag{75}$$

where

$$P^c_{\{n_\alpha\}\{n_{\alpha'}\}}=\sum_{\mathbf{q}\pm}\frac{2\pi}{\hbar}\left(N_{\mathbf{q}}+\frac{1}{2}\pm\frac{1}{2}\right)|H^{\mathbf{q}}_{\{n_\alpha\}\{n_{\alpha'}\}}|^2 \times\delta(\epsilon_{\{n_\alpha\}}-\epsilon_{\{n_{\alpha'}\}}\pm\epsilon_{\mathbf{q}}) \tag{76}$$

are the usual carrier-quasiparticle semiclassical scattering rates given by the well-known Fermi's golden rule. We stress that, contrary to the global (carrier+quasiparticle) description considered in Sec. II E, the scattering rates in Eq. (76) are not symmetric: $P^c_{\{n_\alpha\}\{n_{\alpha'}\}}\neq P^c_{\{n_{\alpha'}\}\{n_\alpha\}}$; this is a direct fingerprint of the irreversible nature of the transport problem induced by energy-relaxation/dephasing processes.

G. Single-particle description

Most of the electronic properties of interest in the analysis of charge-transport phenomena in semiconductor nanostructures are single-particle quantities, i.e., physical quantities ascribed to the generic particle in our electronic subsystem, like carrier drift velocity, mean kinetic energy, etc. In this case, the corresponding quantum-mechanical operator $\hat{A}$ is of the form:

$$\hat{A}=\sum_{\alpha\alpha'}A^{sp}_{\alpha\alpha'}\hat{c}^\dagger_\alpha\hat{c}_{\alpha'} \tag{77}$$

and its average value can be written as

$$A=\sum_{\alpha\alpha'}A^{sp}_{\alpha\alpha'}\mathrm{tr}\{\hat{c}^\dagger_\alpha\hat{c}_{\alpha'}\hat{\rho}\}=\sum_{\alpha\alpha'}A^{sp}_{\alpha\alpha'}\rho^{sp}_{\alpha'\alpha}, \tag{78}$$

where

$$\rho^{sp}_{\alpha_1\alpha_2}=\mathrm{tr}\{\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1}\hat{\rho}\} \tag{79}$$

is the so-called single-particle density matrix.[3] As we can see, this is defined as the average of the product of creation and destruction operators; its diagonal elements ($\alpha_1=\alpha_2$) correspond to the single-particle carrier distribution of the semiclassical Boltzmann theory, while the nondiagonal contributions ($\alpha_1\neq\alpha_2$) describe quantum-mechanical phase coherence between the single-particle states α_1 and α_2. We stress that, while the reduced density-matrix operator $\hat{\rho}^c$ describes the whole many-electron system, the single-particle operator $\hat{\rho}^{sp}$ provides an average or mean-field treatment of the carrier subsystem; indeed the latter fails in describing many-particle correlations, like Coulomb-correlation effects in quasi-zero-dimensional systems.[11]

Since $\rho^{sp}_{\alpha_1\alpha_2}$ is the only quantity entering the evaluation of the average value in Eq. (78), it is desirable to derive a corresponding equation of motion for the single-particle density-matrix in Eq. (79):

$$\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}=\mathrm{tr}\left\{\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1}\frac{d}{dt}\hat{\rho}\right\}. \tag{80}$$

Inserting into the above expression the equation of motion for the global density-matrix operator $\hat{\rho}$ in Eq. (18) we get

$$\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}=\left.\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\right|_{\hat{H}_o}+\left.\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\right|_{\hat{C}}+\left.\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\right|_{\hat{H}'}, \tag{81}$$

where

$$\left.\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\right|_{\hat{H}_o}=\frac{1}{i\hbar}\mathrm{tr}\{\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1}[\hat{H}_o,\hat{\rho}]\} \tag{82}$$

is the time variation induced by the noninteracting Hamiltonian $\hat{H}_o$,

$$\left.\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\right|_{\hat{C}}=-i\,\mathrm{tr}\{\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1}[\hat{\mathcal{H}},\hat{\rho}^i(t)]\} \tag{83}$$

is the contribution due to the quantum-correlation operator $\hat{C}$ in Eq. (22), and

$$\left.\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\right|_{\hat{H}'}=-\mathrm{tr}\{\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1}[\hat{\mathcal{H}},[\hat{\mathcal{K}},\hat{\rho}]]\} \tag{84}$$

is the time evolution dictated by the scattering superoperator Γ.

For a better evaluation of the various contributions in Eqs. (82)–(84) it is convenient to expand the commutators entering the trace, regrouping the various terms in a different way. More specifically, by inserting the explicit form of the single and double commutators, and using the cyclic property of the trace, we finally get

$$\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\bigg|_{\hat{H}_o} = \frac{1}{i\hbar}\mathrm{tr}\{[\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1},\hat{H}_o]\hat{\rho}\}, \tag{85}$$

$$\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\bigg|_{\hat{C}} = -i\,\mathrm{tr}\{[\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1},\hat{\mathcal{H}}]\hat{\rho}^i(t)\}, \tag{86}$$

and

$$\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\bigg|_{\hat{H}'} = -\mathrm{tr}\{[[\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1},\hat{\mathcal{H}}],\hat{\mathcal{K}}]\hat{\rho}\}. \tag{87}$$

As we can see, the various contributions to the time evolution of the single-particle density matrix can be written as global average values of single as well as double commutators.

The term in Eq. (85) can be evaluated exactly. Indeed, recalling the explicit form of the free-carrier+free-phonon Hamiltonian in Eq. (2) and using the Fermionic anticommutation relations $\{\hat{c}_\alpha,\hat{c}^\dagger_{\alpha'}\}=\delta_{\alpha\alpha'}$, we obtain

$$\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\bigg|_{\hat{H}_o} = \frac{\epsilon_{\alpha_1}-\epsilon_{\alpha_2}}{i\hbar}\rho^{sp}_{\alpha_1\alpha_2}. \tag{88}$$

In contrast, for the first- and second-order interaction contributions in Eqs. (86) and (87) it is not possible to obtain closed equations of motion for the single-particle density matrix ρ^{sp}: indeed such contributions involve higher-order correlations, e.g., two-body and/or phonon-assisted density matrices.[3] In order to get a closed equation for $\rho^{sp}_{\alpha_1\alpha_2}$, an additional approximation is needed, the so-called mean-field approximation. The latter consists of a factorization of the higher-order correlation functions into products of single-particle density matrices ρ^{sp} and/or quasiparticle populations $N_{\mathbf{q}}$. The required mean-field procedure and the explicit form of the resulting closed equation of motion depend on the particular form of the interaction Hamiltonian considered, e.g., carrier-carrier, carrier-quasiparticle, etc. However, the free-evolution term (85) together with the first-order contribution in Eq. (86) describe, in general, coherent phenomena—including Hartree-Fock renormalization and coherent phonons—while the second-order term in Eq. (87) describes energy-relaxation/dephasing processes within the Markov approximation.

At this point a few comments are in order. The single-particle description discussed in this section is based on the Schrödinger picture: the equation of motion for $\rho^{sp}_{\alpha_1\alpha_2}$ [see Eq. (80)] is derived by treating the operators $\hat{c}^\dagger$ and $\hat{c}$ in Eq. (79) as time-independent, while the time variation is fully attributed to the density-matrix operator $\hat{\rho}$. Actually, the most popular and commonly used approach[3] to derive the equations of motion governing the time evolution of the single-particle density matrix is based on the Heisenberg picture: the density-matrix operator $\hat{\rho}$ entering Eq. (79) in the Heisenberg scheme is time-independent, while the time evolution is fully ascribed to the Fermionic operators via their corresponding Heisenberg equations of motion:

$$\frac{d}{dt}\hat{c}_\alpha = \frac{1}{i\hbar}[\hat{c}_\alpha,\hat{H}]. \tag{89}$$

More precisely, within the Heisenberg picture Eq. (80) is replaced by

$$\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2} = \mathrm{tr}\left\{\frac{d}{dt}(\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1})\hat{\rho}\right\} = \frac{1}{i\hbar}\mathrm{tr}\{[\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1},\hat{H}]\hat{\rho}\}. \tag{90}$$

Contrary to the theoretical approach proposed in this paper, the usual Heisenberg treatment is based on a correlation expansion of the trace in Eq. (90):[3] starting again from the Hamiltonian separation in Eq. (1), a hierarchy of kinetic equations involving higher-order density as well as quasiparticle-assisted density matrices is established; the different contributions are classified in terms of their perturbation order. Such infinite hierarchy is truncated/closed via the mean-field approximation previously recalled, and only at this level the Markov limit is usually introduced.[3] The aim of this paper, in contrast, is to analyze the Markov limit from a more general point of view; it is for this reason that the latter has been introduced in very general terms in Sec. II C before addressing any reduced description.

1. Carrier-carrier interaction

As the first interaction mechanism we shall consider two-body Coulomb coupling. The corresponding interaction Hamiltonian can be written as

$$\hat{\mathcal{H}}^{cc} = \frac{1}{2\hbar}\sum_{\alpha_1\alpha_2,\alpha_1'\alpha_2'} V^{cc}_{\alpha_1\alpha_2,\alpha_1'\alpha_2'}\hat{c}^\dagger_{\alpha_1}\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1'}\hat{c}_{\alpha_2'}, \tag{91}$$

where $V^{cc}_{\alpha_1\alpha_2,\alpha_1'\alpha_2'}$ is the Coulomb matrix element for the generic two-body transition $\alpha_1'\alpha_2'\to\alpha_1\alpha_2$.

In order to derive the explicit form of the second-order contribution to the single-particle dynamics in Eq. (87), two key quantities need to be evaluated: the inner commutator $[\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1},\hat{\mathcal{H}}]$ and the explicit form of the operator $\hat{\mathcal{K}}$.

More specifically, by employing the anticommutation relations for the Fermionic operators, we get

$$[\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1},\hat{\mathcal{H}}^{cc}] = \frac{1}{\hbar}\sum_{\alpha_3\alpha_4\alpha_5}(V^{cc}_{\alpha_1\alpha_3,\alpha_4\alpha_5}\hat{c}^\dagger_{\alpha_2}\hat{c}^\dagger_{\alpha_3}\hat{c}_{\alpha_4}\hat{c}_{\alpha_5} - V^{cc}_{\alpha_5\alpha_4,\alpha_3\alpha_2}\hat{c}^\dagger_{\alpha_5}\hat{c}^\dagger_{\alpha_4}\hat{c}_{\alpha_3}\hat{c}_{\alpha_1}). \tag{92}$$

The above commutator has the same structure of the interaction Hamiltonian in Eq. (91), i.e., it consists of a sum of products of four Fermionic operators.

By inserting into Eq. (29) the explicit form of the two-body Coulomb interaction Hamiltonian in Eq. (91), we get

$$\hat{\mathcal{K}}^{cc} = \frac{1}{\hbar}\int_0^{t-t_0} d\tau\hat{U}_o(\tau)\left(\frac{1}{2}\sum_{\alpha_1\alpha_2,\alpha_1'\alpha_2'} V^{cc}_{\alpha_1\alpha_2,\alpha_1'\alpha_2'}\hat{c}^\dagger_{\alpha_1}\hat{c}^\dagger_{\alpha_2}\hat{c}_{\alpha_1'}\hat{c}_{\alpha_2'}\right) \times\hat{U}^\dagger_o(\tau). \tag{93}$$

Recalling that

$$\hat{U}_{\rm o}(\tau)\hat{c}^{\dagger}_{\alpha_1}\hat{c}^{\dagger}_{\alpha_2}\hat{c}_{\alpha'_1}\hat{c}_{\alpha'_2}\hat{U}^{\dagger}_{\rm o}(\tau)=\hat{c}^{\dagger}_{\alpha_1}\hat{c}^{\dagger}_{\alpha_2}\hat{c}_{\alpha'_1}\hat{c}_{\alpha'_2}e^{(\epsilon_{\alpha_1}+\epsilon_{\alpha_2}-\epsilon_{\alpha'_1}-\epsilon_{\alpha'_2})\tau/i\hbar}, \tag{94}$$

we finally obtain

$$\hat{\mathcal{K}}^{cc}=\frac{2\pi}{\hbar}\sum_{\alpha_1\alpha_2,\alpha'_1\alpha'_2}\frac{1}{2}V^{cc}_{\alpha_1\alpha_2,\alpha'_1\alpha'_2}\mathcal{D}^{cc}_{\alpha_1\alpha_2,\alpha'_1\alpha'_2}\hat{c}^{\dagger}_{\alpha_1}\hat{c}^{\dagger}_{\alpha_2}\hat{c}_{\alpha'_1}\hat{c}_{\alpha'_2} \tag{95}$$

with

$$\mathcal{D}^{cc}_{\alpha_1\alpha_2,\alpha'_1\alpha'_2}=\frac{1}{2\pi}\int_0^{t-t_0}e^{(\epsilon_{\alpha_1}+\epsilon_{\alpha_2}-\epsilon_{\alpha'_1}-\epsilon_{\alpha'_2})\tau/i\hbar}d\tau. \tag{96}$$

We get again the same operatorial structure: a sum of products of four Fermionic operators.

Given the two results in Eqs. (92) and (95), their commutator—key ingredient in Eq. (87)—will involve, in general, products of six Fermionic operators.

As anticipated, in order to get a closed equation of motion for the single-particle density matrix ρ^{sp} we are forced to employ the mean-field approximation; the latter allows one in this case to write, in general, the average values of six Fermionic operators as products of three single-particle density-matrix elements.

By applying such a mean-field factorization procedure to the explicit form of the outer commutator in Eq. (87), the final result—not reported here—can be cast into the general form:

$$\left.\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\right|_{cc}=\mathcal{F}^{cc,\rm in}[\rho^{sp}]_{\alpha_1\alpha_2}-\mathcal{F}^{cc,\rm out}[\rho^{sp}]_{\alpha_1\alpha_2}. \tag{97}$$

As for the case of the reduced description (see Sec. II F), the time variation of the single-particle density matrix is the sum of a positive—in-scattering—and a negative—out-scattering—contribution. However, contrary to the global and reduced descriptions previously considered, now the in- and out-scattering contributions are nonlinear functions of the single-particle density matrix ρ^{sp}. In particular, in this case of two-body interaction between a main (M) and a partner (P) carrier, the superoperators $\mathcal{F}^{cc,\rm in}$ and $\mathcal{F}^{cc,\rm out}$ both involve a product structure of the form $\hat{\rho}^{sp_{\rm M}}\hat{\rho}^{sp_{\rm P}}(\hat{\mathcal{I}}-\hat{\rho}^{sp_{\rm M}})(\hat{\mathcal{I}}-\hat{\rho}^{sp_{\rm P}})$.

Also for the present single-particle description it is possible to consider the semiclassical or Boltzmann limit introduced in Sec. II E. This amounts again to neglect nondiagonal density-matrix elements: $\rho^{sp}_{\alpha_1\alpha_2}=f^{sp}_{\alpha_1}\delta_{\alpha_1\alpha_2}$. By inserting the above diagonal form of ρ^{sp} into the in- and out-scattering superoperators $\mathcal{F}^{cc,\rm in}$ and $\mathcal{F}^{cc,\rm out}$, the following Boltzmann-like equation for the semiclassical single-particle distribution f^{sp}_{α} may be derived:

$$\frac{df^{sp}_{\alpha}}{dt}=\sum_{\alpha'}[(1-f^{sp}_{\alpha})P^{cc}_{\alpha\alpha'}f^{sp}_{\alpha'}-(1-f^{sp}_{\alpha'})P^{cc}_{\alpha'\alpha}f^{sp}_{\alpha}], \tag{98}$$

where

$$P^{cc}_{\alpha\alpha'}=\frac{2\pi}{\hbar}\sum_{\bar{\alpha}\bar{\alpha}'}(1-f^{sp}_{\bar{\alpha}})|V^{cc}_{\alpha\bar{\alpha},\alpha'\bar{\alpha}'}|^2f^{sp}_{\bar{\alpha}'}\delta(\epsilon_{\alpha}+\epsilon_{\bar{\alpha}}-\epsilon_{\alpha'}-\epsilon_{\bar{\alpha}'}) \tag{99}$$

are two-body carrier-carrier scattering rates describing the main-carrier transition $\alpha'\to\alpha$ accompanied by the partner-carrier transition $\bar{\alpha}'\to\bar{\alpha}$. As we can see, also in the semiclassical limit we deal with a nonlinear transport equation; such nonlinearities are ascribed (i) to the presence of the carrier distribution f^{sp} of the initial partner carrier, and (ii) to the two Pauli-blocking factors $(1-f^{sp})$ corresponding to the final states of both main and partner carriers. Comparing the semiclassical transport equation in Eq. (98) to its quantum-mechanical generalization in Eq. (97), we clearly see that the various terms of the form $(\delta_{\alpha\alpha'}-\rho^{sp}_{\alpha\alpha'})$ are the natural generalization of the Pauli-blocking factors $(1-f^{sp}_{\alpha})$ of the semiclassical theory.

2. Carrier-quasiparticle interaction

Let us now come to the carrier-quasiparticle coupling mechanism. By adopting as explicit form of the carrier-quasiparticle quantity $\hat{\mathcal{H}}_{\mathbf{q}}=\hat{\mathcal{H}}^{\dagger}_{-\mathbf{q}}$ the single-particle operator

$$\hat{\mathcal{H}}_{\mathbf{q}}=\frac{1}{\hbar}\sum_{\alpha\alpha'}g_{\alpha\alpha',\mathbf{q}}\hat{c}^{\dagger}_{\alpha}\hat{c}_{\alpha'}, \tag{100}$$

the carrier-quasiparticle interaction Hamiltonian in Eq. (57) is given by

$$\hbar\hat{\mathcal{H}}^{c\text{-}qp}=\sum_{\alpha\alpha',\mathbf{q}}(g_{\alpha\alpha',\mathbf{q}}\hat{c}^{\dagger}_{\alpha}\hat{b}_{\mathbf{q}}\hat{c}_{\alpha'}+g^{*}_{\alpha\alpha',\mathbf{q}}\hat{c}^{\dagger}_{\alpha'}\hat{b}^{\dagger}_{\mathbf{q}}\hat{c}_{\alpha}), \tag{101}$$

where

$$g_{\alpha\alpha',\mathbf{q}}=\bar{g}_{\mathbf{q}}f_{\alpha\alpha',\mathbf{q}}=\bar{g}_{\mathbf{q}}\int\phi^{*}_{\alpha}(\mathbf{r})e^{i\mathbf{q}\cdot\mathbf{r}}\phi_{\alpha'}(\mathbf{r})d\mathbf{r} \tag{102}$$

is the carrier-quasiparticle matrix element for the single-particle transition $\alpha'\to\alpha$ induced by the quasiparticle bulk mode $\mathbf{q}$. The explicit form of the coupling function $\bar{g}$ depends on the particular carrier-quasiparticle interaction mechanism considered. In any case we have $g_{\alpha\alpha',\mathbf{q}}=g^{*}_{\alpha'\alpha,-\mathbf{q}}$. As for carrier-carrier interaction, in order to derive the second-order contribution to the single-particle dynamics in Eq. (87), we shall evaluate the inner commutator $[\hat{c}^{\dagger}_{\alpha_2}\hat{c}_{\alpha_1},\hat{\mathcal{H}}]$ as well as the operator $\hat{\mathcal{K}}$.

More specifically, by employing again the anticommutation relations for the Fermionic operators we get

$$[\hat{c}^{\dagger}_{\alpha_2}\hat{c}_{\alpha_1},\hat{\mathcal{H}}^{c\text{-}qp}]=-\frac{1}{\hbar}\sum_{\alpha_3,\mathbf{q}}(g_{\alpha_3\alpha_2,\mathbf{q}}\hat{c}^{\dagger}_{\alpha_3}\hat{b}_{\mathbf{q}}\hat{c}_{\alpha_1}+g^{*}_{\alpha_2\alpha_3,\mathbf{q}}\hat{c}^{\dagger}_{\alpha_3}\hat{b}^{\dagger}_{\mathbf{q}}\hat{c}_{\alpha_1}-g_{\alpha_1\alpha_3,\mathbf{q}}\hat{c}^{\dagger}_{\alpha_2}\hat{b}_{\mathbf{q}}\hat{c}_{\alpha_3}-g^{*}_{\alpha_3\alpha_1,\mathbf{q}}\hat{c}^{\dagger}_{\alpha_2}\hat{b}^{\dagger}_{\mathbf{q}}\hat{c}_{\alpha_3}). \tag{103}$$

As we can see, the commutator has the same operatorial structure of the carrier-quasiparticle Hamiltonian, i.e., it involves products of one Bosonic and two Fermionic operators.

By inserting into Eq. (29) the explicit form of the carrier-quasiparticle interaction Hamiltonian in Eq. (101), we get

$$\hat{\mathcal{K}}^{c\text{-}qp} = \frac{1}{\hbar}\int_0^{t-t_0} d\tau \hat{U}_{\rm o}(\tau)\left(\sum_{\alpha\alpha',\mathbf{q}} g_{\alpha\alpha',\mathbf{q}}\hat{c}_\alpha^\dagger \hat{b}_{\mathbf{q}}\hat{c}_{\alpha'}\right)\hat{U}_{\rm o}^\dagger(\tau) + \text{H.c.}, \tag{104}$$

where H.c. denotes its Hermitian conjugate. Recalling that

$$\hat{U}_{\rm o}(\tau)\hat{c}_\alpha^\dagger \hat{b}_{\mathbf{q}}\hat{c}_{\alpha'}\hat{U}_{\rm o}^\dagger(\tau) = \hat{c}_\alpha^\dagger \hat{b}_{\mathbf{q}}\hat{c}_{\alpha'} e^{(\epsilon_\alpha-\epsilon_{\alpha'}-\epsilon_{\mathbf{q}})\tau/i\hbar}, \tag{105}$$

we finally obtain

$$\hat{\mathcal{K}}^{c\text{-}qp} = \frac{2\pi}{\hbar}\sum_{\alpha\alpha',\mathbf{q}} g_{\alpha\alpha',\mathbf{q}}\mathcal{D}^{c\text{-}qp,-}_{\alpha\alpha',\mathbf{q}}\hat{c}_\alpha^\dagger \hat{b}_{\mathbf{q}}\hat{c}_{\alpha'} + \text{H.c.}, \tag{106}$$

with

$$\mathcal{D}^{c\text{-}qp,\pm}_{\alpha\alpha',\mathbf{q}} = \frac{1}{2\pi}\int_0^{t-t_0} e^{(\epsilon_\alpha-\epsilon_{\alpha'}\pm\epsilon_{\mathbf{q}})\tau/i\hbar} d\tau. \tag{107}$$

We get again the same operatorial structure: one Bosonic times two Fermionic operators.

Given the two results in Eqs. (103) and (106), their commutator—key ingredient in Eq. (87)—will involve products of two Bosonic and two Fermionic operators as well as products of four Fermionic operators. As for the carrier-carrier interaction previously discussed, in order to get a closed equation of motion for the single-particle density matrix ρ^{sp} we are forced to employ again the mean-field approximation; the latter allows one to write (i) the average value of two Fermionic times two Bosonic operators as the product of single-particle density-matrix elements $\rho^{sp}_{\alpha\alpha'}$ times quasiparticle distributions, and (ii) the average values of four Fermionic operators as products of two single-particle density-matrix elements.

By applying such a mean-field factorization procedure to the explicit form of the outer commutator in Eq. (87), the final result—not reported here—can be cast in the same form of the one for carrier-carrier interaction in Eq. (97), i.e.,

$$\left.\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\right|_{c\text{-}qp} = \mathcal{F}^{c\text{-}qp,\text{in}}[\rho^{sp}]_{\alpha_1\alpha_2} - \mathcal{F}^{c\text{-}qp,\text{out}}[\rho^{sp}]_{\alpha_1\alpha_2}. \tag{108}$$

As for the case of carrier-carrier interaction, the above in- and out-scattering contributions are again nonlinear functions of the single-particle density matrix ρ^{sp}. More specifically, their general structure is of the form $\hat{\rho}^{sp}(\hat{\mathcal{I}}-\hat{\rho}^{sp})$; such nonlinearities—ascribed to Pauli-blocking effects—vanish in the so-called low-density limit ($\hat{\mathcal{I}}-\hat{\rho}^{sp}\to\hat{\mathcal{I}}$).

In the semiclassical limit, by inserting the diagonal density-matrix form into the in- and out-scattering functionals $\mathcal{F}^{c-qp,\text{in}}$ and $\mathcal{F}^{c-qp,\text{out}}$, we get a nonlinear Boltzmann equation formally identical to the single-particle transport equation in Eq. (98):

$$\frac{df^{sp}_\alpha}{dt} = \sum_{\alpha'}[(1-f^{sp}_\alpha)P^{c\text{-}qp}_{\alpha\alpha'}f^{sp}_{\alpha'} - (1-f^{sp}_{\alpha'})P^{c\text{-}qp}_{\alpha'\alpha}f^{sp}_\alpha], \tag{109}$$

where

$$P^{c\text{-}qp}_{\alpha\alpha'} = \frac{2\pi}{\hbar}\sum_{\mathbf{q}\pm}|g_{\alpha\alpha',\mathbf{q}}|^2\delta(\epsilon_\alpha-\epsilon_{\alpha'}\pm\epsilon_{\mathbf{q}}) \tag{110}$$

denote semiclassical carrier-quasiparticle scattering rates for the single-particle transition $\alpha'\to\alpha$. We deal again with a nonlinear transport equation; in this case, however, such nonlinearities are only ascribed to the Pauli-blocking factor of the final state, and they vanish in the low-density limit as discussed in the following section.

3. Low-density limit

Let us finally consider the so-called "low-density limit." To this aim, let us recall that within the single-particle description previously introduced the average occupation number for the generic state α is simply given by the diagonal elements of the single particle density matrix in Eq. (79):

$$\rho^{sp}_{\alpha\alpha} = \text{tr}\{\hat{c}_\alpha^\dagger\hat{c}_\alpha\hat{\rho}\} = f^{sp}_\alpha. \tag{111}$$

It is then clear that at low carrier concentrations (low densities) the magnitude of all density-matrix elements is much smaller than 1; more precisely, we have $\rho^{sp}_{\alpha\alpha'}\to 0$, $(\delta_{\alpha\alpha'}-\rho^{sp}_{\alpha\alpha'})\to\delta_{\alpha\alpha'}$.

In this limit the carrier-carrier scattering contributions in Eq. (97) vanish since, as anticipated, they involve the single-particle density matrix of the partner carrier $\hat{\rho}^{sp\text{P}}$. As a result, in the low-density limit the only nonvanishing contribution to the interaction dynamics is given by the carrier-quasiparticle terms in Eq. (108). More precisely, by inserting the low-density condition in our quantum transport equation (108) as well as in the corresponding in- and out-scattering functions, we finally get the following linear transport equation:

$$\left.\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2}\right|_{c\text{-}qp} = \sum_{\alpha_1'\alpha_2'}\Gamma_{\alpha_1\alpha_2,\alpha_1'\alpha_2'}\rho^{sp}_{\alpha_1'\alpha_2'} = \sum_{\alpha_1'\alpha_2'}\Gamma^{\text{in}}_{\alpha_1\alpha_2,\alpha_1'\alpha_2'}\rho^{sp}_{\alpha_1'\alpha_2'} - \sum_{\alpha_1'\alpha_2'}\Gamma^{\text{out}}_{\alpha_1\alpha_2,\alpha_1'\alpha_2'}\rho^{sp}_{\alpha_1'\alpha_2'} \tag{112}$$

with

$$\Gamma^{\text{in}}_{\alpha_1\alpha_2,\alpha_1'\alpha_2'} = \frac{2\pi}{\hbar^2}\sum_{\mathbf{q}\pm}\left(N_{\mathbf{q}}+\frac{1}{2}\pm\frac{1}{2}\right) \times(g_{\alpha_1\alpha_1',\mathbf{q}}g^*_{\alpha_2\alpha_2',\mathbf{q}}\mathcal{D}^{c\text{-}qp,\pm *}_{\alpha_2\alpha_2',\mathbf{q}} + \mathcal{D}^{c\text{-}qp,\pm}_{\alpha_1\alpha_1',\mathbf{q}}g_{\alpha_1\alpha_1',\mathbf{q}}g^*_{\alpha_2\alpha_2',\mathbf{q}}) \tag{113}$$

and

$$\Gamma^{\text{out}}_{\alpha_1\alpha_2,\alpha_1'\alpha_2'} = \frac{2\pi}{\hbar^2}\sum_{\alpha'',\mathbf{q}\pm}\left(N_{\mathbf{q}} + \frac{1}{2} \pm \frac{1}{2}\right) \times (\overset{*}{g}_{\alpha''\alpha_1,\mathbf{q}} g_{\alpha''\alpha_1',\mathbf{q}} \mathcal{D}^{c\text{-}qp,\pm}_{\alpha''\alpha_1',\mathbf{q}} \delta_{\alpha_2\alpha_2'} + \delta_{\alpha_1\alpha_1'} \mathcal{D}^{c\text{-}qp,\pm *}_{\alpha''\alpha_2',\mathbf{q}} \overset{*}{g}_{\alpha''\alpha_2',\mathbf{q}} g_{\alpha''\alpha_2,\mathbf{q}}). \quad (114)$$

We stress that the above linear in- and out-scattering operators are formally identical to the reduced-description ones in Eqs. (73) and (74). This can be easily understood, considering that the present low-density limit is physically equivalent to consider a system of just one electron interacting with the quasiparticle degrees of freedom; this would correspond to substitute the many-electron configuration label $\{n_\alpha\}$ with the state α of the only electron considered.

Combining Eqs. (88) and (112) and neglecting the first-order term in Eq. (86), we finally get the following quantum-transport equation:

$$\frac{d}{dt}\rho^{sp}_{\alpha_1\alpha_2} = \mathcal{L}_{\alpha_1\alpha_2,\alpha_1'\alpha_2'}\rho^{sp}_{\alpha_1'\alpha_2'}, \quad (115)$$

where

$$\mathcal{L}_{\alpha_1\alpha_2,\alpha_1'\alpha_2'} = \frac{\epsilon_{\alpha_1} - \epsilon_{\alpha_2}}{i\hbar}\delta_{\alpha_1\alpha_2,\alpha_1'\alpha_2'} + \Gamma_{\alpha_1\alpha_2,\alpha_1'\alpha_2'} \quad (116)$$

is the effective single-particle Liouville operator in the low-density limit.

III. ANALYSIS OF PHASE-COHERENCE EFFECTS IN SEMICONDUCTOR NANOSTRUCTURES

Let us now discuss in more detail the physical meaning of the in- and out-scattering operators in Eqs. (113) and (114). As shown in Eq. (112), the generic matrix element $\Gamma^{\text{in/out}}_{\alpha_1\alpha_2,\alpha_1'\alpha_2'}$ describes how the density-matrix element $\rho_{\alpha_1'\alpha_2'}$ influences the time evolution of the element $\rho_{\alpha_1\alpha_2}$. We shall try to classify such matrix elements according to their role and physical meaning.

As a first class of terms, let us consider the semiclassical ones ($\alpha_1\alpha_1' = \alpha_2\alpha_2'$):

$$\gamma^{T_1}_{\alpha\alpha'} = \Gamma_{\alpha\alpha,\alpha'\alpha'} = P_{\alpha\alpha'} - \delta_{\alpha\alpha'}\sum_{\alpha''} P_{\alpha''\alpha'}. \quad (117)$$

As discussed in the Appendix, the latter, also referred to as T_1 contributions, are fully expressed in terms of the semiclassical rates $P_{\alpha\alpha'}$ [see Eq. (110)] and act on the carrier distribution $f_\alpha = \rho^{sp}_{\alpha\alpha}$ only giving rise to electron and energy redistribution within the traditional Boltzmann picture.

A second class of terms is given by the so-called T_2 contributions ($\alpha_1\alpha_2 = \alpha_1'\alpha_2'$):

$$\gamma^{T_2}_{\alpha_1\alpha_2} = \Gamma_{\alpha_1\alpha_2,\alpha_1\alpha_2} = -\left(\sum_{\alpha''} P_{\alpha''\alpha_1} + \sum_{\alpha''} P_{\alpha''\alpha_2}\right). \quad (118)$$

In the absence of other Γ matrix elements, these T_2 contributions describe the damping of nondiagonal density-matrix elements $p_{\alpha_1\alpha_2} = \rho^{sp}_{\alpha_1\neq\alpha_2}$, also called interstate polarizations;[3] indeed, from Eq. (112) we easily get

$$\left.\frac{d}{dt}p_{\alpha_1\alpha_2}\right|_{c\text{-}qp} = \gamma^{T_2}_{\alpha_1\alpha_2}p_{\alpha_1\alpha_2}, \quad (119)$$

whose solution is simply given by

$$p_{\alpha_1\alpha_2}(t) = e^{\gamma^{T_2}_{\alpha_1\alpha_2}(t-t_0)}p_{\alpha_1\alpha_2}(t_0). \quad (120)$$

The above exponential decay ($\gamma^{T_2}_{\alpha_1\alpha_2}$ is always negative) of nondiagonal density-matrix elements or interstate polarizations is the well-known decoherence or dephasing process.[3] We stress that, in spite of the quantum-mechanical (i.e., coherent) character of $p_{\alpha_1\alpha_2}$, the dephasing rates γ^{T_2} in Eq. (118) involve semiclassical ingredients only; indeed, the latter can be regarded as the sum of the two semiclassical out-scattering rates for states α_1 and α_2 (see the appendix).

In addition to these two classes of terms, acting independently on f_α and $p_{\alpha_1\alpha_2}$, the fully nondiagonal description in Eq. (112) involves also nontrivial coupling contributions, namely (i) coupling between different polarizations ($p_{\alpha_1\alpha_2} \to p_{\alpha_1'\alpha_2'}$), and (ii) terms of the form $\alpha\alpha \to \alpha_1\alpha_2$ and vice versa; the latter describe coupling between f_α and $p_{\alpha_1\alpha_2}$. Denoting with T_3 contributions these last $f \to p$ terms and neglecting $p \to p'$ couplings, the quantum-transport equation in Eq. (112) can be schematically written as

$$\frac{d}{dt}\begin{pmatrix} f \\ p \end{pmatrix} = \begin{pmatrix} \gamma^{T_1} & \gamma^{T_3} \\ \gamma^{T_3} & \gamma^{T_2} \end{pmatrix}\begin{pmatrix} f \\ p \end{pmatrix}, \quad (121)$$

where γ^{T_3} denotes schematically all the $f \to p$ coupling terms previously mentioned.

We clearly see that, in the absence of T_3 terms ($\gamma^{T_3}=0$) there is absolutely no coupling between population (f) and polarization (p). This approximation scheme, known as the T_1T_2 model,[3] is the most popular model used for the interpretation of ultrafast optical experiments in semiconductors.

In contrast, the introduction of these T_3 terms—not included in the conventional T_1T_2 model—gives rise to a nontrivial coupling between f and p, which manifests itself in a residual single-particle phase coherence also in steady-state conditions. More specifically, while within the T_1T_2 model in the limit $t \to \infty$ all polarization terms $p_{\alpha_1\alpha_2}$ vanish [see Eq. (120)] and the resulting steady-state density matrix is diagonal ($\rho^{sp}_{\alpha_1\alpha_2} = f_{\alpha_1}\delta_{\alpha_1\alpha_2}$), in the presence of T_3 terms we may have a steady-state solution characterized by nonzero values of $p_{\alpha_1\alpha_2}$ (see below). As discussed in the appendix in the small-coupling limit the steady-state solution $\rho^{sp}_{\alpha_1\alpha_2}$ of the transport equation in Eq. (115) is always positive-definite. This suggests the introduction of the unitary transformation $T_{\alpha\bar{\alpha}} = \langle\alpha|\bar{\alpha}\rangle$ which diagonalizes the steady-state solution ρ^{sp}. This allows one to clearly express the positive character of the solution according to Eq. (38).

In order to gain more insight into the fully nondiagonal density-matrix approach presented so far, let us consider an extremely simplified scenario: an electronic two-level system interacting with a single phonon mode $\mathbf{q}$. In this case we deal with a two-by-two density matrix of the form:

$$\rho^{sp} = \begin{pmatrix} \rho^{sp}_{aa} & \rho^{sp}_{ab} \\ \rho^{sp}_{ba} & \rho^{sp}_{bb} \end{pmatrix} = \begin{pmatrix} f_a & p^* \\ p & f_b \end{pmatrix}. \tag{122}$$

Here, the diagonal elements ρ^{sp}_{aa} and ρ^{sp}_{bb} coincide with the semiclassical ground—and excited-state level populations f_a and f_b, while the nondiagonal element $p=\rho^{sp}_{ba}$ (together with its complex conjugate $p^*=\rho^{sp}_{ab}$) describes the degree of quantum-mechanical phase coherence between states/levels a and b. Let us introduce the following (arbitrary) $\nu=\{\alpha_1,\alpha_2\}$ mapping: $1=\{a,a\}$, $2=\{b,b\}$, $3=\{b,a\}$, $4=\{a,b\}$. Within such representation, the two-by-two density matrix in Eq. (122) is mapped into a four-dimensional vector, and the single-particle Liouville superoperator $\mathcal{L}$ in Eq. (116) will correspond to a four-by-four matrix. More specifically, within the four-dimensional mapping given above the transport equation (115) in steady-state conditions reduces to the following homogeneous linear problem:

$$\begin{pmatrix} \mathcal{L}_{aa,aa} & \mathcal{L}_{aa,bb} & \mathcal{L}_{aa,ba} & \mathcal{L}_{aa,ab} \\ \mathcal{L}_{bb,aa} & \mathcal{L}_{bb,bb} & \mathcal{L}_{bb,ba} & \mathcal{L}_{bb,ab} \\ \mathcal{L}_{ba,aa} & \mathcal{L}_{ba,bb} & \mathcal{L}_{ba,ba} & \mathcal{L}_{ba,ab} \\ \mathcal{L}_{ab,aa} & \mathcal{L}_{ab,bb} & \mathcal{L}_{ab,ba} & \mathcal{L}_{ab,ab} \end{pmatrix} \begin{pmatrix} f_a \\ f_b \\ p \\ p^* \end{pmatrix} = 0 \tag{123}$$

with

$$\begin{pmatrix} \mathcal{L}_{aa,aa} & \mathcal{L}_{aa,bb} & \mathcal{L}_{aa,ba} & \mathcal{L}_{aa,ab} \\ \mathcal{L}_{bb,aa} & \mathcal{L}_{bb,bb} & \mathcal{L}_{bb,ba} & \mathcal{L}_{bb,ab} \\ \mathcal{L}_{ba,aa} & \mathcal{L}_{ba,bb} & \mathcal{L}_{ba,ba} & \mathcal{L}_{ba,ab} \\ \mathcal{L}_{ab,aa} & \mathcal{L}_{ab,bb} & \mathcal{L}_{ab,ba} & \mathcal{L}_{ab,ab} \end{pmatrix} = \frac{1}{i\hbar} \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & \Delta\epsilon & 0 \\ 0 & 0 & 0 & -\Delta\epsilon \end{pmatrix} + \begin{pmatrix} \Gamma_{aa,aa} & \Gamma_{aa,bb} & \Gamma_{aa,ba} & \Gamma_{aa,ab} \\ \Gamma_{bb,aa} & \Gamma_{bb,bb} & \Gamma_{bb,ba} & \Gamma_{bb,ab} \\ \Gamma_{ba,aa} & \Gamma_{ba,bb} & \Gamma_{ba,ba} & \Gamma_{ba,ab} \\ \Gamma_{ab,aa} & \Gamma_{ab,bb} & \Gamma_{ab,ba} & \Gamma_{ab,ab} \end{pmatrix}. \tag{124}$$

Here $\Delta\epsilon=\epsilon_b-\epsilon_a$ denotes the interlevel energy splitting. Starting from the explicit form of the electron-quasiparticle in- and out-scattering superoperators in Eqs. (113) and (114) and assuming real and symmetric single-particle wave functions $\phi_\alpha(\mathbf{r})$ in Eq. (102), the resulting expression for the matrix elements of Γ for our two-level system is of the form:

$$\begin{pmatrix} \Gamma_{aa,aa} & \Gamma_{aa,bb} & \Gamma_{aa,ba} & \Gamma_{aa,ab} \\ \Gamma_{bb,aa} & \Gamma_{bb,bb} & \Gamma_{bb,ba} & \Gamma_{bb,ab} \\ \Gamma_{ba,aa} & \Gamma_{ba,bb} & \Gamma_{ba,ba} & \Gamma_{ba,ab} \\ \Gamma_{ab,aa} & \Gamma_{ab,bb} & \Gamma_{ab,ba} & \Gamma_{ab,ab} \end{pmatrix} = \frac{\epsilon^{cp}}{\hbar} \begin{pmatrix} -N & (N+1) & 0 & 0 \\ N & -(N+1) & 0 & 0 \\ \frac{i}{2}\xi N & -\frac{i}{2}\xi(N+1) & -\frac{1}{2}(2N+1) & \frac{1}{2}(2N+1) \\ -\frac{i}{2}\xi N & \frac{i}{2}\xi(N+1) & \frac{1}{2}(2N+1) & -\frac{1}{2}(2N+1) \end{pmatrix}, \tag{125}$$

where $\xi=g_{ab,\bar{\mathbf{q}}}(g_{aa,\bar{\mathbf{q}}}-g_{bb,\bar{\mathbf{q}}})/|g_{ab,\bar{\mathbf{q}}}|^2$ (with $\epsilon_{\bar{\mathbf{q}}}=\Delta\epsilon$), ϵ^{cp} denotes the carrier-phonon coupling constant, and N is the Bose occupation number in Eq. (61) for our two-level system.

As already stressed, by neglecting $f\to p$ terms (which correspond to set $g_{aa,\bar{\mathbf{q}}}=g_{bb,\bar{\mathbf{q}}}$), populations and polarizations are totally decoupled. In contrast, in the presence of these T_3 terms our steady-state solution exhibits a residual single-particle coherence, i.e., a polarization p different from zero.

Figure 1 shows the modulus of the interlevel polarization p as a function of the dimensionless coupling parameter $\sigma=\epsilon^{cp}/\Delta\epsilon$. In this numerical example we have chosen $\Delta\epsilon=25$ meV, $T=300$ K, and $a=b=0.25$. As we can see, $|p|$ comes out to be proportional to the coupling parameter σ, as can be readily verified by a closer inspection of our four-by-four superoperator $\mathcal{L}$ in Eq. (124). It follows that for any finite value of the coupling parameter σ, we deal with a nondiagonal steady-state solution ρ [see Eq. (122)]. More specifically, its diagonal elements f_a and f_b are σ-independent and correspond to the semiclassical equilibrium distribution: $f_b/f_a=N/N+1$; the nondiagonal elements p and p^* will increase linearly with the coupling parameter σ. In the small-coupling limit ($\sigma\ll 1$) such nondiagonal contributions are much smaller than the diagonal ones. For increasing values of σ we progressively enter the strong-coupling regime, up to the point where our two-by-two density matrix in Eq. (122) is no more positive definite. Indeed, for $|p|>\sqrt{f_a f_b}$, the determinant of ρ (i.e., $f_a f_b-|p|^2$) becomes negative. As anticipated, this is exactly the regime for which the Markov approximation itself is no longer valid. However, for small and moderate values of σ our two-by-two density matrix is positive-definite, which suggests the introduction of a dressed-state basis in which the latter is diagonal. The new populations $\bar{f}_a$ and $\bar{f}_b$ (dashed curves in Fig. 1) can be regarded as the average occupation of such dressed states. As we can see, for $\sigma=0$ they coincide with the noninteracting thermal ones; for increasing values of the

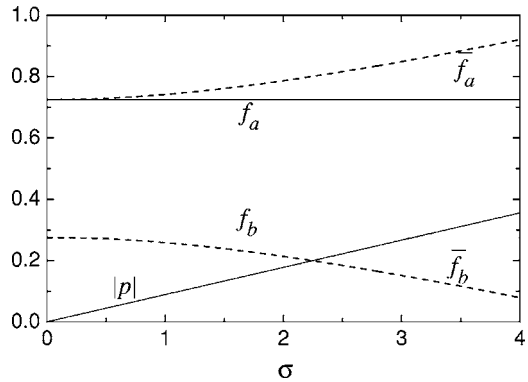

FIG. 1. Phonon-induced single-particle phase coherence for an isolated quantum-dot system: modulus of the interlevel polarization $|p|$ (solid curve) and dressed-states populations $\bar{f}_b$ and $\bar{f}_a$ (dashed curves) as a function of the coupling-constant ratio σ, for an interlevel energy splitting $\Delta\epsilon=25$ meV at room temperature.

carrier-phonon coupling the population ratio $\bar{f}_b/\bar{f}_a$ decreases. Such a behavior can be physically described in terms of a phonon-induced renormalization of the interlevel energy splitting $\Delta\epsilon$. Indeed, such renormalized transition energy can also be obtained from the imaginary parts of the nonzero eigenvalues of the effective Liouville operator in Eq. (124).

We finally stress that such bidimensional dressed basis as well as the corresponding energy shift can be regarded as the simplest example of polaronic phase-coherence and state renormalization (see below).

IV. POLARONIC COHERENCE IN BIASED SEMICONDUCTOR SUPERLATTICES

As a concrete example of scattering-induced single-particle phase coherence in state-of-the-art semiconductor nanostructures, let us consider high-field transport in biased semiconductor superlattices. As extensively discussed in Ref. 27, the treatment of carrier-phonon scattering in the presence of strong electric fields requires a gauge-invariant formulation of the problem; the density-matrix treatment proposed in Ref. 27 has shown that the only single-particle basis α in which the Markov limit is properly defined is the Wannier-Stark one.

In order to gain more insight into the single-particle phase coherence previously discussed, we have performed fully three-dimensional calculations of high-field charge transport in state-of-the-art semiconductor nanostructures. In particular, the aim of our description was to properly treat—in addition to the carrier quantum confinement along the growth direction—the in-plane energy-relaxation and thermalization dynamics.

As a prototypical system, we consider a state-of-the-art GaAs-based nanometric superlattice. More specifically, we have performed a detailed investigation of the 30 Å $GaAs/Al_{0.3}Ga_{0.7}As$ superlattice structure shown in Fig. 2. The single-particle carrier states $\{|\alpha\rangle\}$ are described within the usual envelope-function approximation in terms of a

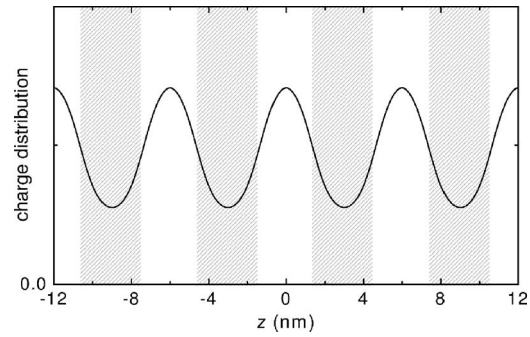

FIG. 2. Schematics of the prototypical 30 Å/30 Å $GaAs/Al_{0.3}Ga_{0.7}As$ superlattice structure considered in our simulated experiments: Real-space periodic nanostructure profile (shaded regions correspond to barriers) and charge distribution corresponding to the ground-state ($k_\parallel=0$) envelope function in the field-free case $[(n(r_\parallel)\propto|\phi^0_{k_\parallel=0,\nu=1}(r_\parallel)|^2)]$.

space-independent effective mass m^*. They come out to be products of two-dimensional plane waves and one-dimensional envelope functions:

$$\phi_\alpha(\mathbf{r})=\frac{1}{\sqrt{\Omega}}e^{i\mathbf{k}_\perp\cdot\mathbf{r}_\perp}\phi^\parallel_{\alpha_\parallel}(r_\parallel), \tag{126}$$

Ω denoting a suitable normalization area.

In the free-field case, the envelope functions $\phi^\parallel$ in Eq. (126) reduce to one-dimensional Bloch states $\phi^\parallel_{k_\parallel\nu}$, corresponding to the periodic heterostructure potential reported in the inset of Fig. 3. As we can see, we deal with a relatively small band-edge discontinuity ($V_o=250$ meV). The latter, combined with a barrier width of 30 Å, gives rise to significant interwell carrier tunneling. This is confirmed by the field-free ground-state charge distribution (solid curve in Fig. 2), which shows a clear fingerprint of carrier delocalization.

The interwell coupling displayed in Fig. 2 should translate into a dispersive energy-momentum relation along the growth direction. This is confirmed by the superlattice miniband profile reported in Fig. 3. As we can see, we deal with a carrier miniband only: Its width amounts to about 90 meV

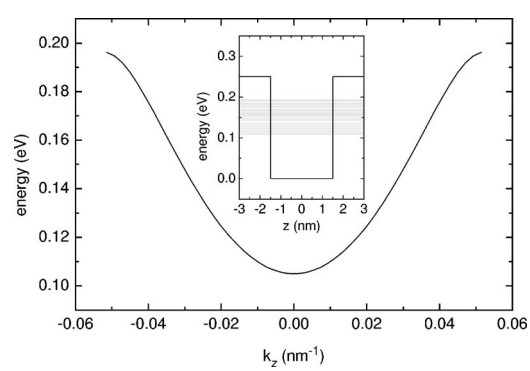

FIG. 3. Single-miniband diagram (miniband width of about 90 meV) corresponding to the superlattice structure depicted in Fig. 2. The superlattice potential profile (band-edge discontinuity of 0.25 eV) is also shown in the inset.

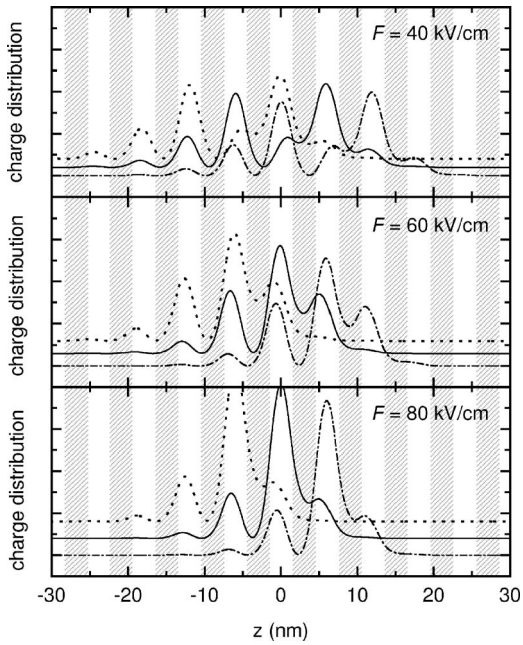

FIG. 4. Charge distribution corresponding to the Wannier-Stark states in the superlattice structure of Fig. 2 for different values of the applied field F. For each field three states are displayed: $n=-1$ (dotted curve), $n=0$ (solid curve), and $n=1$ (dash-dotted curve).

and it is therefore larger than the longitudinal optical (LO)-phonon energy ($\simeq$36 meV).

In the presence of an applied field F along the growth direction, the one-dimensional envelope functions $\phi^{\parallel}$ within the scalar-potential gauge correspond to the usual Wannier-Stark states.[27] The latter are displayed in Fig. 4 for different values of F. As we can see, for increasing values of the field we deal with an increasing state localization and a corresponding suppression of interwell single-particle tunneling.

The primary goal of our simulated experiments was the study of the current-voltage characteristics of the superlattice structure previously introduced, in the presence of carrier-LO phonon scattering. In particular, a thermal bath of three-dimensional (bulk) dispersionless longitudinal polar optical phonons has been assumed, employing the carrier-phonon interaction model given in Ref. 28. We have therefore evaluated the carrier drift velocity as a function of the applied field. The latter can be readily computed according to Eq. (78), using as the single particle quantity the velocity operator

$$\mathbf{A}^{sp} = \mathbf{v} = \frac{\mathbf{P}}{m^*} = -\frac{i\hbar}{m^*}\nabla . \tag{127}$$

More specifically, we get

$$v^d = \sum_{\alpha\alpha'} v_{\alpha\alpha'}\rho^{sp}_{\alpha'\alpha}, \tag{128}$$

where $\rho^{sp}_{\alpha\alpha'}$ is the single-particle density matrix, and $v_{\alpha\alpha'}$ denote the matrix elements of the velocity operator (127) within our α representation.

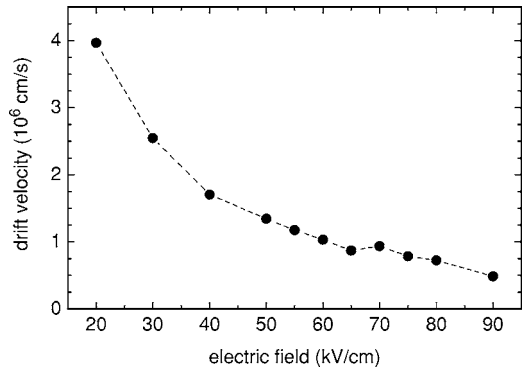

FIG. 5. Steady-state carrier drift velocity as a function of the applied field for the superlattice structure of Fig. 2 at room temperature, in the low-density limit. The line is a guide to the eye.

Figure 5 shows the steady-state carrier drift velocity as a function of the applied field for the superlattice structure of Fig. 2 at room temperature in the low-density limit. The peak at $\simeq$70 kV/cm corresponds to the phonon resonance, i.e., for this value of the applied field, the Wannier-Stark energy eFd—d being the superlattice period—is equal to the LO-phonon energy. We stress that such a feature agrees well with the results obtained with the nonequilibrium Green's function treatment by Jauho and co-workers.[29] As we can see, we get drift-velocity values significantly different from zero. This remark is particularly important in view of the fact that such field-induced current is entirely due to the presence of nondiagonal density-matrix elements; indeed, within the semiclassical limit previously introduced we have $\rho^{sp}_{\alpha\alpha'}=f^{sp}_{\alpha}\delta_{\alpha\alpha'}$, and therefore Eq. (128) reduces to

$$v^d = \sum_{\alpha} v_{\alpha\alpha} f^{sp}_{\alpha}. \tag{129}$$

As we can see, the only matrix elements of the velocity operator involved are the diagonal ones, i.e., $v_{\alpha=\alpha'}$; for localized as well as periodic basis states α such diagonal matrix elements are known to vanish, and therefore within the semiclassical limit the drift velocity in Eq. (128) is equal to zero, i.e., no current.

It is then easy to conclude that the significantly large values of the drift velocity reported in Fig. 5 are entirely due to the nondiagonal density-matrix elements $\rho^{sp}_{\alpha\neq\alpha'}$ entering Eq. (128); in turn such nondiagonal contributions reflect a scattering induced phase coherence between our Wannier-Stark states. As a result, we can say that the significant carrier drift velocity in Fig. 5 is an unambiguous fingerprint of phonon-induced phase coherence in semiconductor superlattices.

V. SUMMARY AND CONCLUSIONS

We have presented a general density-matrix formulation of quantum-transport phenomena in semiconductor nanostructures typically employed in the design of state-of-the-art quantum devices. More specifically, contrary to the conventional single-particle correlation expansion, we have investi-

gated separately the effects of the Markov limit and of the reduction procedure. Our fully operatorial approach has allowed us to better identify the general properties of the scattering superoperators entering our effective quantum-transport theory at various description levels, e.g., N electrons-plus-quasiparticles, N electrons only, and single-particle picture.

In addition to coherent transport phenomena characterizing the transient response of the system, the proposed theoretical description has allowed the study of scattering induced phase coherence in steady-state conditions. In particular, based on the proposed approach we have investigated polaronic effects in strongly biased semiconductor superlattices.

One important conclusion of our theoretical analysis is that the Markov limit alone—i.e., not combined with the diagonal or semiclassical approximation—may lead to totally nonphysical results: (i) within the global (carrier +quasiparticle) picture the Markov limit introduces a fictitious energy redistribution/relaxation and dephasing which has no physical counterpart within a closed-system scenario; (ii) both for the global and for the reduced pictures, the resulting effective Liouville superoperators do not correspond, in general, to a CP map, thus preventing any "robust" time-dependent solution of our quantum-transport equation.

A second important remark is that the combination of the Markov limit with the semiclassical approximation—i.e., the neglect of nondiagonal density-matrix elements—leads, also for the global description, to Boltzmann-like equations whose solution can be shown to be positive-definite at any time.

In spite of the fact that our density-matrix formulation within the usual Markov limit does not translate into CP maps, our analysis has shown that within the small-coupling limit the proposed quantum-transport equations are always characterized by a positive-definite steady-state solution.

ACKNOWLEDGMENTS

We are grateful to Massimo Fischetti, Carlo Jacoboni, Tilmann Kuhn, and Paolo Zanardi for stimulating and fruitful discussions.

APPENDIX: GENERAL PROPERTIES OF THE SCATTERING SUPEROPERATOR

The aim of this Appendix is to discuss the general structure and symmetry properties which characterize the global (carrier+quasiparticle) in- and out-scattering superoperators in Eqs. (33) and (34), their reduced versions in Eqs. (73) and (74), and the single-particle ones in Eqs. (113) and (114).

To this end, let us consider first the semiclassical limit introduced in Sec. II E. For the global picture as well as for the reduced and single-particle descriptions we deal with a Boltzmann transport equation of the form

$$\frac{df_i}{dt} = \sum_{i'} (P_{ii'} f_{i'} - P_{i'i} f_i). \tag{A1}$$

In analogy with the definition of the scattering superoperator Γ in Eq. (26), the transport equation (A1) can be rewritten as

$$\frac{df_i}{dt} = \sum_{i'} \gamma_{ii'} f_{i'}, \tag{A2}$$

where

$$\gamma_{ii'} = \gamma_{ii'}^{\text{in}} - \gamma_{ii'}^{\text{out}} = P_{ii'} - \delta_{ii'} \sum_{i''} P_{i''i} \tag{A3}$$

is again the difference of in- and out-contributions. As a first remark, we notice that (i) the in-contribution coincides with the semiclassical scattering rate $P_{ii'}$ and (ii) the out-contribution is a function of the in-one:

$$\gamma_{ii'}^{\text{out}} = \delta_{ii'} \sum_{i''} \gamma_{i''i}^{\text{in}}. \tag{A4}$$

The Boltzmann transport equation in Eq. (A1) is supposed to preserve the total number of particles $N=\Sigma_i f_i$, i.e.,

$$\frac{dN}{dt} = \sum_i \frac{df_i}{dt} = \sum_{ii'} \gamma_{ii'} f_{i'} = 0. \tag{A5}$$

The above conservation law—which corresponds to the trace-preserving property of the quantum-mechanical case—needs to be verified for any carrier distribution f_i; this requires that

$$\sum_i \gamma_{ii'} = 0, \tag{A6}$$

which can be readily verified starting from the definition of the operator γ in Eq. (A3).

Let us now consider the eigenvalue problem for the semiclassical scattering operator γ:

$$\sum_{i'} \gamma_{ii'} f_{i'} = \Lambda f_i. \tag{A7}$$

As a first property, the eigenvalue spectrum is nonpositive definite: $\Lambda \leqslant 0$; second, the particle-preserving property in Eq. (A6) requires the existence of the $\Lambda=0$ eigenvalue, whose eigenvector f_i corresponds to the steady-state solution of the problem.

We shall finally discuss the positive-definite character of the semiclassical carrier dynamics. Starting from the Boltzmann equation in Eq. (A2), we can express the carrier distribution at time t in terms of the semiclassical propagator $s_{ij}(t-t_0)$:

$$f_i(t) = \sum_j s_{ij}(t-t_0) f_j(t_0), \tag{A8}$$

where $s_{ij}(0)=\delta_{ij}$ and

$$\frac{ds_{ij}}{dt} = \sum_{i'} \gamma_{ii'} s_{i'j}. \tag{A9}$$

By inserting into the above equation the explicit form of the scattering operator γ in Eq. (A3) we get

$$\frac{ds_{ij}}{dt} = -\bar{\gamma}_i s_{ij} + \sum_{i'} P_{ii'} s_{i'j}, \tag{A10}$$

where $\bar{\gamma}_i = \Sigma_{i'} P_{i'i}$ denotes the total or out-scattering rate for state i. The above differential equation can be formally integrated according to

$$s_{ij}(t) = e^{-\bar{\gamma}_i(t-t_0)}\delta_{ij} + \int_{t_0}^{t} dt' e^{-\bar{\gamma}_i(t-t')} \sum_{i'} P_{ii'} s_{i'j}(t'). \tag{A11}$$

This formal solution—also known as Chamber's formulation[30]—is the starting point of the semiclassical Neumann expansion. Indeed by iteratively substituting Eq. (A11) into itself, we obtain

$$s_{ij}(t) = \sum_{n=0}^{\infty} \int_{t_0}^{t} dt_1 \cdots \int_{t_0}^{t_{n-1}} dt_n \sum_{i_1,\ldots,i_{n-1}} e^{-\bar{\gamma}_i(t-t_1)} \times P_{ii_1} e^{-\bar{\gamma}_{i_1}(t_1-t_2)} \cdots P_{i_{n-1}j} e^{-\bar{\gamma}_j(t_n-t_0)}. \tag{A12}$$

The above expansion can be regarded as the semiclassical counterpart of the Neumann series in Eq. (15).[30] By recalling that the scattering rates $P_{ii'}$ and $\bar{\gamma}_i$ are positive-definite quantities, it is easy to recognize that all the terms of the above Neumann expansion are non-negative (i.e., greater or equal to zero); therefore we can conclude that all the matrix elements $s_{ij}(t)$ of the semiclassical propagator are positive-definite, which in turn ensures that the distribution function f_i in Eq. (A8) is itself positive-definite. As anticipated, we see that the combination of the Markov limit with the semiclassical or diagonal approximation leads to a completely positive time evolution, both for the global (carrier +quasiparticle) case and for the reduced as well as single-particle pictures.

Let us now move to the quantum-mechanical case. A closer inspection of the in- and out-scattering superoperators in Eqs. (33) and (34) as well as of their reduced versions in Eqs. (73) and (74) and (113) and (114) reveals that they have a general structure of the form

$$\Gamma^{\text{in}}_{i_1 i_2, i_1' i_2'} = \mathcal{P}^{\text{in}}_{i_1 i_2, i_1' i_2'} + \mathcal{P}^{\text{in}*}_{i_2 i_1, i_2' i_1'}, \tag{A13}$$

$$\Gamma^{\text{out}}_{i_1 i_2, i_1' i_2'} = \mathcal{P}^{\text{out}}_{i_1 i_2, i_1' i_2'} + \mathcal{P}^{\text{out}*}_{i_2 i_1, i_2' i_1'}, \tag{A14}$$

with

$$\mathcal{P}^{\text{in}}_{i_1 i_2, i_1' i_2'} = \frac{2\pi}{\hbar^2} g_{i_1 i_1'} g^*_{i_2 i_2'} \mathcal{D}^*_{i_2 i_2'} \tag{A15}$$

and

$$\mathcal{P}^{\text{out}}_{i_1 i_2, i_1' i_2'} = \delta_{i_2 i_2'} \sum_{i''} \mathcal{P}^{\text{in}*}_{i'' i'', i_1 i_1'}. \tag{A16}$$

Exactly as in the semiclassical theory previously recalled [see Eq. (A4)], the out-scattering superoperator is fully determined by the in-one. The latter, in turn, can be regarded as the quantum-mechanical (i.e., nondiagonal) generalization of the conventional Fermi's golden rule; indeed, its semiclassical contributions ($i_1 i_1' = i_2 i_2'$) are of the form

$$\Gamma^{\text{in}}_{ii, i'i'} = \frac{2\pi}{\hbar^2} |g_{ii'}|^2 (\mathcal{D}_{ii'} + \mathcal{D}_{i'i}). \tag{A17}$$

Similar to the particle-conservation law in Eq. (A5), in the quantum-mechanical case we ask the scattering superoperator

$$\Gamma_{i_1 i_2, i_1' i_2'} = \Gamma^{\text{in}}_{i_1 i_2, i_1' i_2'} - \Gamma^{\text{out}}_{i_1 i_2, i_1' i_2'} \tag{A18}$$

to be trace preserving, i.e.,

$$\frac{d}{dt} \text{tr}\{\hat{\rho}\} = \text{tr}\{\Gamma(\hat{\rho})\} = 0. \tag{A19}$$

Rewriting the above equation within our generic i-basis we get

$$\sum_i \sum_{i_1' i_2'} \Gamma_{ii, i_1' i_2'} \rho_{i_1' i_2'} = 0. \tag{A20}$$

This relation should hold for any generic ρ; this requires that

$$\sum_i \Gamma_{ii, i_1' i_2'} = 0. \tag{A21}$$

The above trace-preserving condition can be readily verified starting from the general structure of the in- and out-scattering superoperators in Eqs. (A13) and (A14). We stress that such property is verified for any form of the in-scattering superoperator $\mathcal{P}^{\text{in}}$, i.e., does not depend on the specific structure of the scattering operator in Eq. (A15).

Let us now come to the eigenvalue problem for the scattering superoperator Γ and for the corresponding effective Liouville superoperator $\mathcal{L}$ [see Eqs. (115) and (116)]. By denoting with $j = i_1 i_2$ the generic pair of indices for our density matrix, we have

$$\mathcal{L}_{jj'} \rho_{j'} = \Lambda \rho_j. \tag{A22}$$

Contrary to the semiclassical picture, now the eigenvalues Λ are complex quantities and, more importantly their real parts may assume positive as well as negative values. This is a clear indication of the fact that we are not dealing with a CP map (see below). However, the trace-preserving property in Eq. (A21) requires also for the quantum-mechanical case the existence of the $\Lambda = 0$ eigenvalue, whose eigenvector $\rho_j = \rho_{i_1 i_2}$ corresponds to the steady-state solution of the problem.

We shall finally discuss the possible positive-definite character of our density-matrix ρ. In very general terms we can express the density matrix at time t via a quantum-mechanical propagator or quantum-mechanical map $\mathcal{S}$:

$$\rho_{i_1 i_2}(t) = \sum_{j_1 j_2} \mathcal{S}_{i_1 i_2, j_1 j_2}(t - t_0)\rho_{j_1 j_2}(t_0), \tag{A23}$$

where $\mathcal{S}_{i_1 i_2, j_1 j_2}(0) = \delta_{i_1 i_2, j_1 j_2}$ and

$$\frac{d}{dt}\mathcal{S}_{i_1 i_2, j_1 j_2} = \sum_{i_1' i_2'} \mathcal{L}_{i_1 i_2, i_1' i_2'}\mathcal{S}_{i_1' i_2', j_1 j_2}. \tag{A24}$$

Using the more compact operatorial notation we have

$$\hat{\rho}(t) = \mathcal{S}(\hat{\rho}(t_0)) \tag{A25}$$

with

$$\frac{d\mathcal{S}}{dt} = \mathcal{L}\mathcal{S}. \tag{A26}$$

It is well-known that the most general form of a CP map is given by

$$\mathcal{S}(\hat{\rho}(t_0)) = \sum_k \hat{\mathcal{M}}_k(t - t_0)\hat{\rho}(t_0)\hat{\mathcal{M}}^\dagger(t - t_0), \tag{A27}$$

where the generic set of operators $\hat{\mathcal{M}}_k$—usually referred to as "Krauss operators"—should obey the normalization condition

$$\sum_k \hat{\mathcal{M}}_k \hat{\mathcal{M}}_k^\dagger = \hat{\mathcal{I}}. \tag{A28}$$

A particular class of CP maps is given by the so-called Lindblad maps, whose effective Liouville superoperators are of the form:

$$\mathcal{L}(\hat{\rho}) = \frac{1}{i\hbar}[\hat{H}_o, \hat{\rho}] + \sum_k ([\hat{\mathcal{A}}_k \hat{\rho}, \hat{\mathcal{A}}_k^\dagger] + [\hat{\mathcal{A}}_k, \hat{\rho}\hat{\mathcal{A}}_k^\dagger]). \tag{A29}$$

For the particular case of Hermitian operators $\hat{\mathcal{A}}_k$, the Lindblad form in Eq. (24) is recovered.

Let us now try to compare the effective scattering dynamics described by the transport equation in Eq. (115) with the general Krauss expansion in Eq. (A27) and with the Lindblad form in Eq. (A29).

For the global description, the double-commutator in Eq. (19) is not Lindblad-like; indeed, as already pointed out, its eigenvalue spectrum is in general not negative-definite. This tells us that, while the steady-state solution of the Lindblad form in Eq. (24) is again proportional to the identity operator $\hat{\mathcal{I}}$, its dynamical evolution in general does not preserve the positive-definite character of $\hat{\rho}$ and may also exhibit singularities. Such anomalous and totally non-physical behavior—not present within the semiclassical or Boltzmann picture—is again the result of the Markov limit.

Also for the case of the reduced and single-particle descriptions, their effective Liouville operators derived in Secs. II F and II G do not correspond, in general, to CP maps, and its time evolution does not preserve the positive-definite character of $\hat{\rho}$.

In spite of the fact that, in general, we are not dealing with CP maps, it is possible to show that for all the transport equations derived in the paper we have a steady-state solution. Moreover, in the so-called small-coupling limit, i.e., when the perturbation Hamiltonian $\hat{H}'$ is small compared to the noninteracting Hamiltonian $\hat{H}_o$, it is possible to show that such a steady-state $(\Lambda = 0)$ solution is always positive-definite [see Eq. (38)].

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