

Relaxation properties of the quantum kinetics of carrier-LO-phonon interaction in quantum wells and quantum dots

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The time evolution of optically excited carriers in semiconductor quantum wells and quantum dots is analyzed for their interaction with LO phonons. Both the full two-time Green's function formalism and the one-time approximation provided by the generalized Kadanoff-Baym ansatz are considered, in order to compare their description of relaxation processes. It is shown that the two-time quantum kinetics leads to thermalization in all the examined cases, which is not the case for the one-time approach in the intermediate-coupling regime, even though it provides convergence to a steady state. The thermalization criterion used is the Kubo-Martin-Schwinger condition.

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I. INTRODUCTION

The Boltzmann equation is of central importance in many fields of physics and, since its original formulation in the theory of gases it has received a whole range of extensions to other domains like plasma physics, nuclear physics, or semiconductors. In these fields, Boltzmann scattering integrals are extensively used to model relaxation and thermalization processes. Adapted versions of the H theorem ensure that, indeed, the equations describe the steady evolution of the system towards the proper thermal equilibrium. Early attempts to derive this irreversible behavior from the quantum-mechanical evolution have shown¹ that the range of validity of Boltzmann-like equations correspond to the low-coupling, slowly varying, long-time regime.

In more recent years with the experimental possibility to produce and control transport and optical phenomena at ultrashort time scales, quantum-kinetic theories^{2,3} have been devised in order to describe rapid processes in which coherence is still present, together with the onset of dephasing and relaxation. This means that the kinetics has to describe not only real-number quantities like occupation probabilities, but also complex, off-diagonal density-matrix elements, and their interference effects.

Not only fast dynamics, but also the necessity to extend the theory beyond the weak-interaction limit has prompted the development of quantum kinetics. A typical example is provided by the interaction of carriers with LO phonons in semiconductor quantum dots, where a phonon bottleneck is predicted by the Boltzmann result (see Ref. 4, and references therein), in contrast to the quantum-kinetic treatment of quantum-dot polarons in the strong-coupling regime⁵ and many experimental findings.

The quantum-kinetic theory using nonequilibrium Green's functions (GF) is one of the basic tools in this field. Its central object is the one-particle, two-time GF, for which closed equations are provided. Unfortunately, the large numerical effort needed for solving these equations has limited previous applications of the two-time formalism to the early-time regime. The method has been used to describe the ultrafast

optical excitation of semiconductors where the interaction of carriers with LO phonons,⁶ the Coulomb interaction of carriers,^{7,8} and their combined influence⁹ have been studied. Calculations based on the two-time formalism also have been applied in plasma physics¹⁰ and for nuclear matter.¹¹

Since the physically relevant information (e.g., population and polarization dynamics) is contained in the one-time GF (the two-time GF at equal times), it is clear that a closed equation for this quantity would greatly simplify the procedure. This explains the huge popularity of the generalized Kadanoff-Baym ansatz (GKBA),¹² an approximation which expresses the two-time GF in terms of its one-time component.

The GKBA has been extensively used in the past for a description of non-Markovian contributions to ultrafast relaxation and dephasing processes. Signatures of non-Markovian effects have been investigated for the interaction of carriers with LO phonons.^{13,14} Furthermore, the buildup of screening has been studied on the basis of a quantum-kinetic description using the GKBA (Refs. 15 and 16) and included in scattering calculations.^{17,18} Results of the one-time and two-time formulation have been compared for early times addressing the carrier-carrier scattering¹⁰ as well as the interaction of carriers with LO phonons.¹⁹

Boltzmann-like kinetic equations are obtained from the one-time theory based on the GKBA by further approximations: memory effects are neglected (Markov limit) and free particle energies are used (low coupling limit). One encounters therefore a situation in which only after taking two major approximation steps, one reaches a kinetic theory for which the physically expected correct relaxation behavior can be proven analytically. To our knowledge, there is no attempt in the literature to explore systematically the relaxation properties of either the two-time formalism or its one-time approximation, despite their wide applications and the obvious fundamental importance of the problem. For example, the interest in laser devices based on quantum wells²⁰ and quantum dots²¹ requires a good understanding of the long-time behavior of the carriers in their evolution to equilibrium. Furthermore, the importance of non-Markovian ef-

fects in the quantum-kinetic treatment of optical gain spectra for quantum-dot lasers has been discussed recently.^{22,23}

In this paper, the relaxation properties in the long-time limit are compared for the one-time and two-time quantum kinetics. As a test case, we consider the interaction of carriers with LO phonons in semiconductor nanostructures, which is the dominant relaxation mechanism for low carrier densities and elevated temperatures. We study the optical excitation of quantum wells and quantum dots with short laser pulses and calculate the dephasing of the coherent polarization together with the relaxation and thermalization of the excited carrier populations. The equilibrium state of the interacting system is defined by the Kubo-Martin-Schwinger condition. We investigate if and under which conditions this equilibrium state is reached in the time-dependent solution of the quantum kinetic models. This provides a unique way to address the range of validity of the involved approximations.

II. RELAXATION PROPERTIES OF THE BOLTZMANN EQUATION

The Markovian limit of the kinetics, as described by the Boltzmann equation, is a good example to start with, because its relaxation properties are well understood and rigorously proven. To be specific, we consider the Hamiltonian for the interacting system of carriers and phonons

$$H_{\text{e-ph}} = \sum_i \epsilon_i a_i^\dagger a_i + \sum_q \hbar \omega_q b_q^\dagger b_q + \sum_{i,j,q} M_{i,j}(\mathbf{q}) a_i^\dagger a_j (b_q + b_{-q}^\dagger), \quad (1)$$

where i, j are indices for the carrier states and the momentum \mathbf{q} is the phononic quantum number. The corresponding creation and annihilation operators for carriers and phonons are given by a_i^\dagger, a_i and b_q^\dagger, b_q , respectively. The Boltzmann equation for the time evolution of the average occupation number (population distribution) $f_i = \langle a_i^\dagger a_i \rangle$ has the form

$$\frac{\partial f_i}{\partial t} = \sum_j [W_{i,j}(1-f_i)f_j - W_{j,i}(1-f_j)f_i], \quad (2)$$

with the transition rates given by Fermi's golden rule

$$W_{i,j} = \frac{2\pi}{\hbar} \sum_q |M_{i,j}(\mathbf{q})|^2 [N_q \delta(\epsilon_i - \epsilon_j - \hbar \omega_q) + (N_q + 1) \delta(\epsilon_i - \epsilon_j + \hbar \omega_q)]. \quad (3)$$

For a phonon bath in thermal equilibrium, N_q is a Bose-Einstein distribution with the lattice temperature, and the δ functions ensure the strict energy conservation in the $j \rightarrow i$ transition process assisted by either the absorption or the emission of a phonon.

The following properties of Eq. (2) can be analytically proven: (i) the total number of carriers $\sum_i f_i$ is conserved, (ii) positivity is preserved, i.e., if at $t=0$ one has $f_i \geq 0$ then this remains true at any later time, (iii) the Fermi distribution $f_i = [e^{-\beta(\epsilon_i - \mu)} + 1]^{-1}$ is a steady-state solution of Eq. (2), and (iv) this steady state is the large time limit of the solution $f_i(t)$ for any positive initial condition *provided* a certain con-

nectivity property holds. This property is fulfilled if any state of the carrier system can be reached from any other state through a chain of transitions having nonzero rates. The temperature of the stationary Fermi distribution is the lattice temperature, and the chemical potential is fixed by the total number of carriers. If the set of carrier states is not connected in the above sense, any connected component behaves like a separate fluid and reaches equilibrium with its own chemical potential.

As satisfying as this picture looks, several problems arise here. The carrier-phonon interaction is essential as a relaxation mechanism but the carrier energies themselves are taken as if unaffected by it. Both in the energy conserving δ functions and in the final Fermi distribution these energies appear as unperturbed. This corresponds to a low-coupling regime, which may not be valid in practical situations. Even in weakly polar semiconductors like GaAs, the confined nature of the states in quantum wells (QWs) and even more so in quantum dots (QDs), gives rise to an enhanced effective interaction.²⁴ For higher coupling constants one expects departures from the simple picture discussed above. Moreover, in the case of a strong coupling and with the inclusion of memory effects, neglected in the Markovian limit, the energy conservation is not expected to hold. Finally, and specifically for LO phonons, their dispersionless spectrum, associated with strict energy conservation turns the system into a disconnected one. Indeed, each carrier can move only up and down a ladder with fixed steps of size $\hbar \omega_{LO}$ but cannot jump on states outside this ladder. A phonon bottleneck effect in QDs was predicted on these grounds.⁴

III. STATEMENT OF THE PROBLEM

It is clear that in most practical cases one has to turn to quantum-kinetic treatments in which both energy renormalizations and memory effects are considered. Such formalisms are provided by the two-time Green's function kinetics or by one-time approximations to it. In view of the discussion of the previous section, the following questions, regarding the relaxation properties of the quantum kinetics, are in order: (i) Is the particle number conserved? (ii) Is positivity conserved? (iii) Is the system evolving to a steady state? (iv) If yes, is this steady state a thermal equilibrium one? In what sense?

To our knowledge, with the exception of the first question, which can be easily answered affirmatively, there is no definite and proven answer available in the literature. The aim of the present paper is to investigate how numerical solutions of the quantum-kinetic equations for realistic situations behave in the discussed respects. For this purpose, we compare the results of the two-time and the one-time approach.

IV. TWO-TIME QUANTUM KINETICS

In this section we specify the Hamiltonian, Eq. (1), for the case of a homogeneous two-band semiconductor, where carriers interact with LO phonons via the Fröhlich coupling,

$$H_{e-ph} = \sum_{k,\lambda} \epsilon_k^\lambda a_{k,\lambda}^\dagger a_{k,\lambda} + \sum_q \hbar \omega_q b_q^\dagger b_q + \sum_{k,q,\lambda} g_q a_{k+q,\lambda}^\dagger a_{k,\lambda} (b_q + b_{-q}^\dagger). \quad (4)$$

The carrier quantum numbers are the band index $\lambda=c,v$ and the three-dimensional (3D)—(for the bulk case) or two-dimensional (2D)—(for QWs) momentum \mathbf{k} . The coupling is defined by $g_q^2 \sim \alpha/q^2$ for the 3D case, or by $g_q^2 \sim \alpha F(q)/q$ for the quasi-2D case, with the form factor $F(q)$ related to the QW confinement function and the Fröhlich coupling constant α . Additional terms to this Hamiltonian describe the optical excitation and the Coulomb interaction in the usual way.³ We consider only sufficiently low excitations so that carrier-carrier scattering and screening effects are negligible. Then the only contribution of the Coulomb interaction is the Hartree-Fock renormalization of the single particle energies and of the Rabi frequency.

The object of the kinetic equations is the two-time GF, $G_k^{\lambda,\lambda'}(t_1, t_2) = G_k^{\lambda,\lambda'}(t, t - \tau)$. We use the parametrization of the two-time plane (t_1, t_2) in terms of the main time t and relative time τ . One can combine the two Kadanoff-Baym equations²⁵ which give the derivatives of the GF with respect to t_1 and t_2 , according to $\partial/\partial t = \partial/\partial t_1 + \partial/\partial t_2$ and $\partial/\partial \tau = -\partial/\partial t_2$ in order to propagate the solution either along the time diagonal (t equation) or away from it (τ equation). As two independent GFs we choose the lesser and the retarded ones, and limit ourselves to the subdiagonal halfplane $\tau \geq 0$, since supradiagonal quantities can be related to subdiagonal ones by complex conjugation. With these options and in matrix notation with respect to band indices, the main-time equation reads

$$i\hbar \frac{\partial}{\partial t} G_k^{R,<}(t, t - \tau) = \Sigma_k^{R,<}(t) G_k^{R,<}(t, t - \tau) - G_k^{R,<}(t, t - \tau) \Sigma_k^{\delta}(t - \tau) + i\hbar \left. \frac{\partial}{\partial t} G_k^{R,<}(t, t - \tau) \right|_{\text{coll}}, \quad (5)$$

where the instantaneous self-energy contains the external and the self-consistent field

$$\Sigma_k^{\delta}(t) = \begin{pmatrix} \epsilon_k^c & -\hbar \Omega_R(t) \\ -\hbar \Omega_R^*(t) & \epsilon_k^v \end{pmatrix} + i\hbar \sum_q V_q G_{k-q}^{<}(t, t). \quad (6)$$

The collision term in Eq. (5) has different expressions for G^R and $G^{<}$,

$$i\hbar \left. \frac{\partial}{\partial t} G_k^R(t, t - \tau) \right|_{\text{coll}} = \int_{t-\tau}^t dt' [\Sigma_k^R(t, t') G_k^R(t', t - \tau) - G_k^R(t, t') \Sigma_k^R(t', t - \tau)], \quad (7)$$

$$i\hbar \left. \frac{\partial}{\partial t} G_k^{<}(t, t - \tau) \right|_{\text{coll}} = \int_{-\infty}^t dt' [\Sigma_k^R G_k^{<} + \Sigma_k^{<} G_k^A - G_k^R \Sigma_k^{<} - G_k^{<} \Sigma_k^A]. \quad (8)$$

The time arguments of the self-energies and GFs in Eq. (8)

are the same as in Eq. (7) and are omitted for simplicity. The advanced quantities are expressible through retarded ones by conjugation. The self-energies are computed in the self-consistent random phase approximation (RPA) scheme and have the explicit expressions

$$\begin{aligned} \Sigma_k^R(t, t') &= i\hbar \sum_q g_q^2 [D_q^>(t - t') G_{k-q}^R(t, t') \\ &\quad + D_q^R(t - t') G_{k-q}^{<}(t, t')], \\ \Sigma_k^{<}(t, t') &= i\hbar \sum_q g_q^2 D_q^{<}(t - t') G_{k-q}^{<}(t, t'), \end{aligned} \quad (9)$$

with the equilibrium phonon propagator

$$D_q^{\geq}(t) = -\frac{i}{\hbar} [N_q e^{\pm i\hbar \omega_q t} + (1 + N_q) e^{\mp i\hbar \omega_q t}]. \quad (10)$$

For practical calculations, we use dispersionless phonons $\omega_q = \omega_{LO}$.

The above set of equations has to be supplemented by specifying the initial conditions. For all the times prior to the arrival of the optical pulse, the system consists of the electron-hole vacuum in the presence of the phonon bath. This is an equilibrium situation, characterized by diagonal GFs, which depend only on the relative time. More precisely, one has

$$\begin{aligned} G^R(t, t - \tau) &= \begin{pmatrix} g_c^R(\tau) & 0 \\ 0 & g_v^R(\tau) \end{pmatrix}, \\ G^{<}(t, t - \tau) &= \begin{pmatrix} 0 & 0 \\ 0 & -g_v^R(\tau) \end{pmatrix}, \end{aligned} \quad (11)$$

where g^R is the retarded GF of the unexcited system. The calculation of this GF is an equilibrium problem, namely that of the Fröhlich *polaron*. The polaronic GF is the solution of the τ equation

$$\left(i\hbar \frac{\partial}{\partial \tau} - \epsilon_k^\lambda \right) g_{k,\lambda}^R(\tau) = \int_0^\tau d\tau' \Sigma_{k,\lambda}^R(\tau - \tau') g_{k,\lambda}^R(\tau'), \quad (12)$$

in which, to be consistent with the t evolution described above, the RPA self-energy is again used. The vacuum GF of Eq. (11) is not only the starting value for the GF in the main-time evolution but it also appears in the integrals over the past which require GF values before the arrival of the optical pulse. Moreover, the presence of the polaronic GF brings into the picture the complexities of the spectral features of the polaron, with energy renormalization and phonon satellites. Finally, the decay of the polaronic GF introduces a natural memory depth into the problem. An example is seen in Fig. 1 where, due to a rather strong coupling constant and a high temperature, the decay with the relative time τ is rapid. This allows to cut the infinite time integrals of Eq. (8) at a certain distance away from the diagonal.

In Fig. 1 the momentum argument is replaced by the unrenormalized electron energy $E = \hbar^2 k^2 / 2m_e^*$. This change of variable is allowed by the fact that the momentum dependence of the polaronic GF is isotropic. The same energy

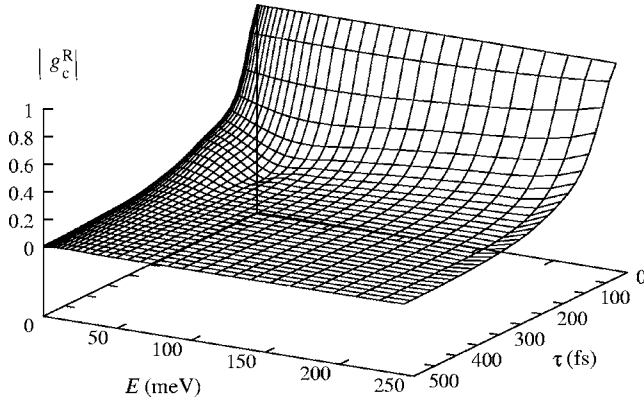


FIG. 1. Absolute value of the polaronic retarded GF for electrons in a CdTe QW at $T=300$ K. $E=0$ corresponds to the conduction band edge.

argument is used in the subsequent figures, with the exception of Fig. 2 where the reduced mass is employed ($E = \hbar^2 k^2 / 2m_r^*$) as being more appropriate in the case of the polarization. The choice of an energy variable facilitates the comparison with other energies appearing in the theory. For instance, in Fig. 1 a somewhat slower decay is seen at low energies and is a trace of the phonon threshold ($\hbar\omega_{LO} = 21$ meV). Also, in Figs. 2 and 3 below, the functions have a peak around the detuning (120 meV).

With the specification of the initial conditions, the problem to be solved is completely defined. After obtaining the two-time GFs, the physically relevant information is found in the equal-time lesser GF, which contains the carrier populations and the polarization. This program is carried out for the case of a CdTe ($\alpha=0.31$) QW at room temperature. The excitation conditions are defined by a Gaussian-shaped pulse of 100 fs duration [full width at half maximum (FWHM) of the intensity], having an excess energy of 120 meV above the unrenormalized band gap and an area of 0.05 (in π units). This gives rise to carrier densities in the order of $10^9/\text{cm}^2$, sufficiently low to neglect carrier-carrier scattering.

As seen in Figs. 2 and 3, the interaction of carriers with LO phonons provides an efficient dephasing and leads, in a subpicosecond time interval, to a relaxation of the electron population into a steady-state distribution. The same is true for the hole population (not shown). Before discussing this result we compare it to the outcome of the one-time calculation.

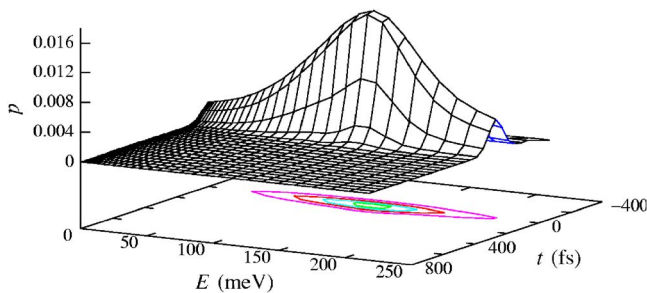


FIG. 2. (Color online) Time evolution of the coherent interband polarization after optical excitation of a CdTe QW with a 100 fs pulse centered at time $t=0$, using a two-time calculation.

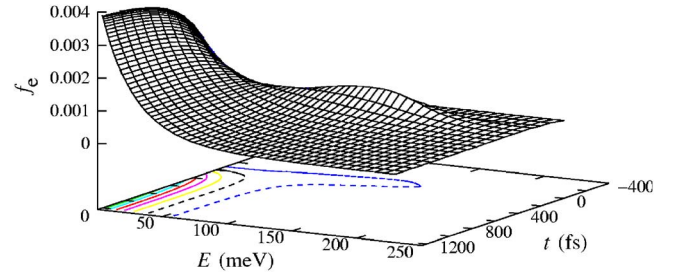


FIG. 3. (Color online) Time evolution of the electron population distribution for the same situation as in Fig. 2.

V. ONE-TIME QUANTUM KINETICS

To obtain from the Kadanoff-Baym equations for the two-time GF a closed set of equations for the equal-time lesser GF, $G^<(t, t)$, one can use the GKBA.¹² The ansatz reduces the time-offdiagonal GFs appearing in the collision terms of Eq. (8) to diagonal ones with the help of the spectral (retarded or advanced) GFs. Therefore, the GKBA has to be supplemented with a choice of spectral GFs. In our case, it is natural to use for this purpose the polaronic GF. For $\tau \geq 0$, this leads to the GKBA in the form

$$G^<(t, t - \tau) \approx i\hbar g^R(\tau) G^<(t - \tau, t - \tau). \quad (13)$$

The result of this procedure for the same system and using the same excitation conditions as for the two-time calculation is shown in Fig. 4. We find that the steady state obtained in this way differs appreciably from that of the two-time calculation.

VI. THE KUBO-MARTIN-SCHWINGER CONDITION

For a fermionic system in thermal equilibrium, the following relationship connects the lesser and the spectral GF (Ref. 2)

$$G_k^<(\omega) = -2if(\omega)\text{Im} G_k^R(\omega),$$

$$f(\omega) = \frac{1}{e^{\beta(\hbar\omega - \mu)} + 1}. \quad (14)$$

In thermodynamic equilibrium, the GFs depend only on the relative time and Eq. (14) involves their Fourier transform with respect to this time. The relationship is known as the

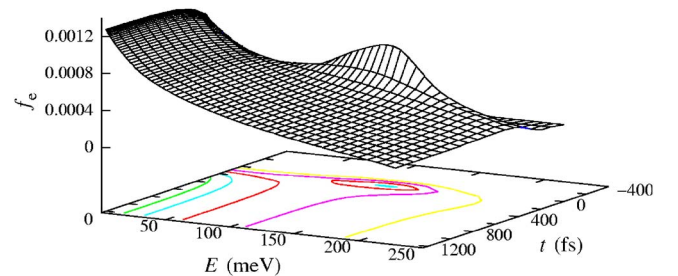


FIG. 4. (Color online) Time evolution of the electron population distributions in a CdTe QW, using the same excitation conditions as in Figs. 2 and 3 and a one-time calculation.

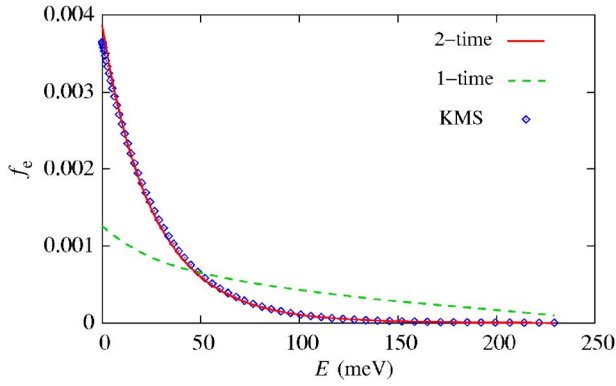


FIG. 5. (Color online) One-time and two-time CdTe QW electron populations at $t=1240$ fs and the KMS result.

Kubo-Martin-Schwinger (KMS) condition or as the fluctuation-dissipation theorem, and leads to a thermal equilibrium population given by

$$f_k^\lambda = - \int \frac{d\hbar\omega}{\pi} f(\omega) \text{Im} G_k^{R,\lambda\lambda}(\omega). \quad (15)$$

The two-time theory provides the excitation-dependent retarded GF along with the lesser one, the formalism being a system of coupled equation for these two quantities. Nevertheless, in the low excitation regime used here the difference between the actual retarded GF and its vacuum counterpart $g_{k,\lambda}^R(\omega)$ turns out to be negligible, as can be checked numerically. Therefore, the latter can be used in Eq. (15) without loss of accuracy.

The thermal equilibrium distribution f_k^λ obtained from the KMS condition is the generalization of the Fermi function of the noninteracting case and is used as a check of a proper thermalization. The test of the two steady-state solutions against the KMS distribution function is seen in Fig. 5. The two-time calculation is in good agreement with the KMS curve, but the one-time evolution is not. It appears that the one-time kinetics produces a steady state with a temperature considerably exceeding that of the phonon bath.

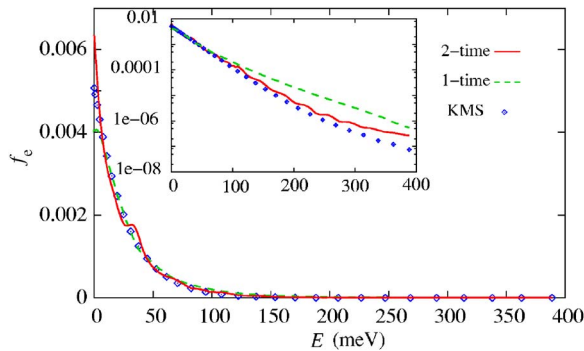


FIG. 6. (Color online) Electron population at $t=1600$ fs for a GaAs QW and optical excitation with a 100 fs laser pulse at $t=0$. Solutions of the two-time and the one-time quantum kinetics are compared with the KMS result. Inset: same on semilogarithmic scale.

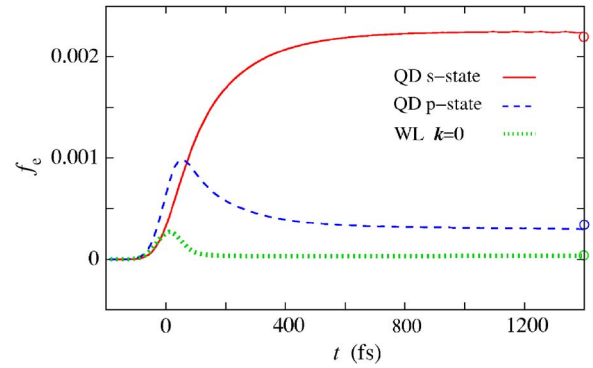


FIG. 7. (Color online) Electron populations in the localized s and p states of a CdTe QD and in the extended $\mathbf{k}=0$ WL state after optical excitation with a 100 fs laser pulse at $t=0$, as calculated using the two-time kinetics. Open circles represent the equilibrium values according to the KMS condition.

It is to be expected, however, that for a weaker coupling the discrepancy between the full two-time procedure and the GKBA is less severe. This is indeed the case, as shown in Fig. 6, where results for a GaAs ($\alpha=0.069$) QW are given. The wiggles seen in the two-time curve are traces of the phonon cascade, which are still present. This is due to the much longer relaxation time in low-coupling materials. Nevertheless the trend is clear, the steady-state solutions of both approaches are in good agreement with the KMS condition.

Another important example concerns a non-homogeneous system. It consists of CdTe lens-shaped self-assembled QDs, having both for electrons and for holes two discrete levels below the wetting-layer (WL) continuum. These states are labeled s and p , according to their z -projection angular momentum. We consider an equidistant energy spacing of $2.4\hbar\omega_{LO}$ between the WL continuum edge, the p level and the s level, for the electrons and a $0.27\hbar\omega_{LO}$ similar spacing for holes. The formalism used is the same as for the homogeneous systems but with the momentum replaced by a state quantum number running over the discrete QD states and the WL continuum. This amounts to neglecting GF matrix elements which are off-diagonal in the state index, but still keeping off-diagonal terms with respect to the band index. This has been shown to be a reasonable approximation for

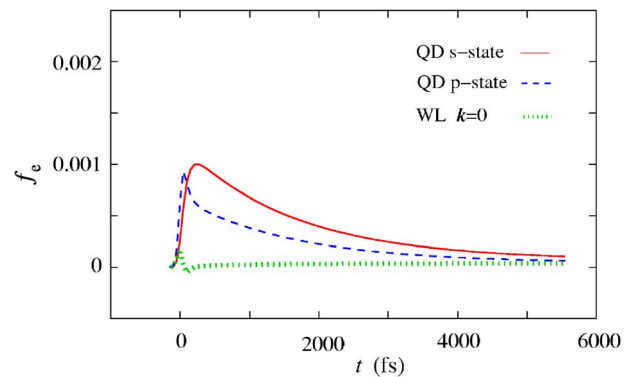


FIG. 8. (Color online) Same as Fig. 7, but using the one-time kinetics. Note that an identical ordinate axis is used to facilitate the comparison.

QDs.^{5,26} Our calculations for this example include both localized QD and delocalized WL states. We consider a harmonic in-plane confinement potential for the localized states and construct orthogonalized plane waves for the delocalized states in the WL plane. The strong confinement in growth direction is described by a steplike finite-height potential. Details for the calculation of interaction matrix elements are given in Ref. 5. In Figs. 7 and 8, the time evolution of the population of electrons is shown. The system is pumped close to the renormalized p -shell energy with a 100 fs laser pulse at time $t=0$. Therefore, the majority of the carriers is initially found in the p state (which has a twofold degeneracy due to the angular momentum in addition to the spin degeneracy). Nevertheless, efficient carrier relaxation takes place, even if the level spacing does not match the LO-phonon energy, and a steady state is reached. The two-time results are again in agreement with the KMS condition, shown by open circles. The one-time evolution shows a non-physical intermediate negative value for the WL population and converges to a state in strong disagreement with the KMS result.

VII. CONCLUSIONS

The long-time behavior of different quantum kinetic approaches to the problem of carrier scattering by means of LO phonons was analyzed, in order to assess their relaxation properties. As a test of proper convergence to thermal equilibrium, the KMS condition was used. We considered materials with low (GaAs) and intermediate (CdTe) Fröhlich coupling. The results can be summarized as follows: (i) In both the one-time and the two-time quantum kinetics steady states are reached. (ii) The steady state produced by the two-time approach obeys the KMS condition in all cases considered. (iii) The one-time result agrees with the KMS condition only at low coupling and differs considerably for larger ones.

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