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

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Abstract.
We compare numerically the carrier-LO-phonon quantum kinetics as provided by the two-time Green’s function formalism on the one hand and its one-time approximation given by the generalized Kadanoff-Baym ansatz on the other hand. Self-consistent RPA self-energies are used in both cases. The aim of the analysis is to check the correct thermalization behaviour of the solutions. It is shown that both formalisms give rise to steady states in the long time limit. It is shown that these states correspond to the correct thermal equilibrium for the two-time solutions, while the one-time approach fails to thermalize the carriers in the intermediate-coupling regime. The thermalization criterion used is the KMS condition.

1. Introduction
The scattering of carriers with LO-phonon is the dominant relaxation mechanism at low excitation densities. Information on dephasing and scattering in the early stages after the optical excitation can be obtained in pump-probe experiments. These data were successfully modelled within the frame of quantum kinetics [1, 2, 3]. On the other hand, the interest for laser devices based on quantum wells [4] and quantum dots [5] requires a good understanding of the long-time behaviour of the carriers in their evolution to equilibrium. In this paper different kinetic approaches are studied comparatively from the point of view of their relaxation properties. Since these are all, to a smaller or greater extent, approximations to the exact theory, it is by no means granted that important features are not lost. The analysis of such features provides numerical arguments about the range of validity of these approximations.

2. Relaxation properties of the Boltzmann equation
The markovian limit of the kinetics, as described by the Boltzmann equation, is sufficiently simple to allow analytical proof of its relaxation properties, which are therefore quite well understood. To be specific, we consider the carrier-phonon Hamiltonian

\[ H_{e-ph} = \sum_i \epsilon_i a_i^\dagger a_i + \sum_q \hbar \omega q b_q^\dagger b_q + \sum_{i,j,q} M_{i,j}(q) a_i^\dagger a_j (b_q + b_{-q}^\dagger) \]  (1)

where \( i, j \) are indices for the carrier states and the momentum \( q \) is the phononic quantum number. The Boltzmann equation for the time evolution of the average occupation number
(population distribution) \( f_i = \langle a_i^\dagger a_i \rangle \) reads

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

with the transition rates given by Fermi’s Golden Rule

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

For a phonon bath in equilibrium \( N_q \) is a Bose-Einstein distribution with the lattice temperature, and the \( \delta \)-functions ensure the strict energy conservation in the \( j \rightarrow i \) transition process involving 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 \( f_i = \left[ e^{-\beta(\epsilon_i - \mu)} + 1 \right]^{-1} \) 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 connectivity property holds. This property holds if any state of the carrier system can be reached from any other state through a chain of transitions having non-zero rates. The temperature of the limiting Fermi distribution is then 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.

These satisfactory properties notwithstanding, several problems arise here. The carrier-phonon interaction is essential for the relaxation but its influence on the energy levels is ignored. Both in the energy conserving \( \delta \)-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 confinement of the states in quantum wells (QWs) and in quantum dots (QDs), is enhancing the effective interaction [6]. Under these conditions 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.

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 included. Such formalisms are provided by the two-time Green’s function (GF) 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: Is the particle number conserved? Is positivity conserved? Is the system evolving to a steady state? 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 provide some numerical evidence in this problem, by studying comparatively the results of the two-time and the one-time approach on several examples.

3. Two-time quantum kinetics

In this section we follow closely the two-time formalism described in a previous paper [7]. We specify the Hamiltonian of Eq.(1) for the case of a homogeneous two-band semiconductor, with carriers interacting with LO-phonons via the Fröhlich coupling, as
\[ H_{e-ph} = \sum_{k,\lambda} c_k^\dagger a_{k,\lambda} a_{k,\lambda} + \sum_q \hbar \omega_q b_q^\dagger b_q + \sum_{k,\tilde{q},\lambda} g_q a_{k+\tilde{q},\lambda}^\dagger a_{k,\lambda} (b_q + b_{-q}^\dagger). \] (3)

The carrier quantum numbers are the band index \( \lambda = c, v \) and the 3D- (for the bulk case) or 2D- (for QWs) momentum \( \vec{k} \). 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 \( F(q) \) the form factor related to the QW confinement function and \( \alpha \) the adimensional Fröhlich coupling constant. Additional terms to this Hamiltonian describe the optical excitation and the Coulomb interaction in the usual way. We consider only sufficiently low excitations so that carrier-carrier scattering and screening effects are negligible, the only contribution of the Coulomb term being the Hartree-Fock renormalization of the energies and of the Rabi frequency.

The object of the kinetic equations is the two-time GF, \( G^\lambda_{\vec{k}}(t_1, t_2) = G^\lambda_{\vec{k}}(t, t - \tau) \). We use the parametrization of the two-time plane \((t_1, t_2)\) in terms of the main time \( t \) and relative time \( \tau \). One can combine the two Kadanoff-Baym equations [8] 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 (\( \tau \)-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

\[
\left. i\hbar \frac{\partial}{\partial t} G^{\lambda, \lambda'}_{\vec{k}}(t, t - \tau) \right|_{\text{coll}} = \Sigma^\rho_{\vec{k}}(t) G^{\lambda, \lambda'}_{\vec{k}}(t, t - \tau) - G^{\rho, \lambda'}_{\vec{k}}(t, t - \tau) \Sigma^\rho_{\vec{k}}(t - \tau) + \left. i\hbar \frac{\partial}{\partial t} G^{\lambda, \lambda'}_{\vec{k}}(t, t - \tau) \right|_{\text{coll}},
\] (4)

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

\[
\Sigma^\rho_{\vec{k}}(t) = \begin{pmatrix}
\epsilon^c_{\vec{k}} & -\hbar \Omega^c_{\vec{k}}(t) \\
-\hbar \Omega^c_{\vec{k}}(t) & \epsilon^v_{\vec{k}}
\end{pmatrix} + i\hbar \sum_q V_q \left. G^{\lambda, \lambda'}_{\vec{k}-q}(t, t) \right|_{\text{coll}}.
\] (5)

The collision term in Eq.(4) has different expression for \( G^R \) and \( G^L \)

\[
\left. i\hbar \frac{\partial}{\partial t} G^R_{\vec{k}}(t, t - \tau) \right|_{\text{coll}} = \int_{-\tau}^t dt' \left[ \Sigma^R_{\vec{k}}(t, t') G^R_{\vec{k}}(t', t - \tau) - G^R_{\vec{k}}(t, t') \Sigma^R_{\vec{k}}(t', t - \tau) \right],
\] (6)

\[
\left. i\hbar \frac{\partial}{\partial t} G^L_{\vec{k}}(t, t - \tau) \right|_{\text{coll}} = \int_{-\infty}^t dt' \left[ \Sigma^L_{\vec{k}} G^L_{\vec{k}}(t, t') + \Sigma^L_{\vec{k}} G^A_{\vec{k}} - G^R_{\vec{k}} \Sigma^L_{\vec{k}} - G^L_{\vec{k}} \Sigma^A_{\vec{k}} \right].
\] (7)

The time arguments of the self-energy and GF in Eq.(7) are the same as in Eq.(6) and are omitted for simplicity. The advanced quantities are expressible through retarded ones by conjugation. The self-energy is computed in the self-consistent RPA scheme and has the explicit expression

\[
\Sigma^R_{\vec{k}}(t, t') = i\hbar \sum_q g_q^2 \left[ D^R_q(t - t') G^R_{\vec{k}-q}(t, t') + D^L_q(t - t') G^L_{\vec{k}-q}(t, t') \right],
\]

\[
\Sigma^L_{\vec{k}}(t, t') = i\hbar \sum_q g_q^2 D^L_q(t - t') G^L_{\vec{k}-q}(t, t'),
\] (8)
with $D_q(t)$ the equilibrium phononic propagator.

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

$$
G^R(t, t - \tau) = \begin{pmatrix} g^R_c(\tau) & 0 \\ 0 & g^R_v(\tau) \end{pmatrix}; \quad G^<(t, t - \tau) = \begin{pmatrix} 0 & 0 \\ 0 & -g^R_c(\tau) \end{pmatrix},
$$

(9)

where $g^R$ is the retarded GF of the empty system. The calculation of this GF is an equilibrium problem, namely that of the Fröhlich polaron. The polaronic GF is the solution of the $\tau$-equation

$$
\left( i\hbar \frac{\partial}{\partial \tau} - \epsilon^\lambda_k \right) g^R_{k,\lambda}(\tau) = \int_0^\tau d\tau' \Sigma^R_{k,\lambda}(\tau - \tau') g^R_{k,\lambda}(\tau'),
$$

(10)
in which, to be consistent with the $t$-evolution described above, the RPA self-energy is again used. The vacuum GF of Eq.(9) 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 $\tau$ is rapid. This allows to cut the infinite time integrals of Eq.(7) at a certain distance away from the diagonal. (In all the figures the energy is the unrenormalized electron parabolic band $E = \hbar^2 k^2 / 2m_e^*$ with the exception of Fig. 2, where the reduced mass is used $E = \hbar^2 k^2 / 2m_r^*$.)

With the specification of the initial conditions the problem to be solved is completely defined. After finding 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 (FWHM of the intensity), having an excess energy of 120meV above the unrenormalized band gap and an area of 0.05 (in $\pi$ units). This gives rise to carrier densities of the order of $10^9$/$\text{cm}^2$, sufficiently low to neglect carrier-carrier scattering.

As seen in Figs. 2, 3, the carrier-LO-phonon interaction provides an efficient dephasing and leads, in a sub-picosecond 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.
4. One-time quantum kinetics

As it is well known, one reaches a closed set of equations for the equal-time lesser GF $G^<(t, t)$ by making use of the generalized Kadanoff-Baym ansatz (GKBA)\cite{9}. The ansatz reduces the time-offdiagonal GFs appearing in the collision terms of Eq(7) to diagonal ones with the help of the spectral (retarded or advanced) GFs. Therefore, in practice 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. This leads to the GKBA in the form

$$G^<(t, t - \tau) \approx i \hbar \, g^R(\tau) \, G^<(t - \tau, t - \tau).$$ \hspace{1cm} (11)

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. It is clear that the steady state obtained in this way differs appreciably from that of the two-time calculation.
5. The KMS condition

For a fermionic system in thermal equilibrium, the following relationship connects the lesser and the spectral GF [1]

\[ G^<_{\vec{k}}(\omega) = -2i f(\omega) \text{Im} G^R_{\vec{k}}(\omega); \quad f(\omega) = \frac{1}{e^{\beta(\bar{\hbar}\omega - \mu)} + 1}. \]  

(12)

In equilibrium the GFs depend only on the relative time and Eq.(12) involves their Fourier transform with respect to this time. The relationship is known as the Kubo-Martin-Schwinger (KMS) condition or as the fluctuation-dissipation theorem. Using again the polaronic retarded GF, the thermal equilibrium populations are given by

\[ f^\lambda_{\vec{k}} = -\int \frac{d\hbar\omega}{\pi} f(\omega) \text{Im} g^R_{\vec{k},\lambda}(\omega). \]  

(13)

The distribution obtained from the KMS condition is the generalization of the Fermi function of the non-interacting case and is used as a check of a proper thermalization.

The test of the two steady-state solutions against the KMS condition 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 final state which is hotter than the phonon bath.

![Figure 5. One- and two-time CdTe QW electron populations at t = 1240 fs and the KMS result.](image)

It is to be expected, however, that for a lower 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, both approaches being in good agreement with the KMS condition.

6. Conclusions

The relaxation behaviour of QW carriers under LO-phonon scattering was numerically analyzed both in the two-time GF formalism and in its one-time (GKBA) approximation. Both low (GaAs) and intermediate (CdTe) Fröhlich couplings were considered. It was shown that in all cases steady states are reached. This is interesting in itself, since no analytic argument is known for proving the convergence to a steady state in these kinetic theories. On the other hand, not all steady states are necessarily states of thermal equilibrium. The criterion for proper thermalization is the KMS condition. We have shown that the two-time formalism leads to thermalization in all cases considered, while the one-time approximation fails in this respect for stronger couplings.
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Figure 6. One- and two-time GaAs QW electron populations at $t = 1600$ fs and the KMS result. Inset: the same in semilogarithmic plot.